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Volume XIV

TECHNICAL PAPERS FROM THE LENINGRAD INTERNATIONAL SYMPOSIUM ON THE METEOROLOGICAL ASPECTS OF ATMOSPHERIC POLLUTION

PART III

Edited By

M. Y. Nuttonson

The material presented here is part of a survey of
USSR literature on air pollution
conducted by the Air Pollution Section
AMERICAN INSTITUTE OF CROP ECOLOGY

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PREFACE

The present volume constitutes Part III* of translations of papers presented in Leningrad during July 1968 at the International Symposium on Meteorological Aspects of Air Pollution. The original papers delivered at the Symposium were edited by Prof. M. E. Berlyand and published as a report in 1971 by the Hydrometeorological Publishing House in Leningrad.

The report contains a total of 40 papers, 37 of which are in Russian and three, in German, together with their accompanying abstracts. In this volume we present a collection of translations of the remaining papers from the original report.

A review article by T. A. Ogneva, "International Symposium on Meteorological Aspects of Atmospheric Pollution" (Izv. VGO**, Vol. 101, No. 4, 1969, pp. 395-396) provides interesting background material regarding the Symposium. For this reason we present our translation of Ogneva's paper in the following paragraphs.

"At the present time, considerable attention is being given to the study of atmospheric pollution in order to improve the sanitary status of water and air reservoirs in urban areas and workers' settlements and to intensify measures aimed at the preservation of nature. In working out the measures designed to decrease the pollution of air by noxious impurities, a special role is played by the consideration of meteorological factors, that substantially determine the behavior (dispersal) of impurities in the atmosphere. This problem was the subject of an International Symposium on Meteorological Aspects of Atmospheric Pollution, held in Leningrad at the A. I. Voeykov Main Geophysical Observatory on 22-31 July 1968. The organizers of the Symposium were the State Committee of the USSR Council of Ministers on Science and Technology (Scientific Council on the Problem 'Protection of the Air Reservoir from Pollution by Noxious Substances') and the Main Administration of the Hydrometeorological Service, Council of Ministers of the USSR.

"The Symposium was attended by scientists and specialists (meteorologists, hygienists, chemists, power engineers, metallurgists, etc.) from Bulgaria, Hungary, Vietnam, the German Democratic Republic, Poland, Rumania, Czechoslovakia, and the Soviet Union. Forty-five reports were delivered, and there was an animated discussion on the subjects of physical principles and methods of calculation of dispersal of industrial emissions in the atmosphere; meteorological parameters determining atmospheric diffusion and intensity of atmospheric pollution; procedures and equipment for observing atmospheric pollution, and experimental studies of pollution of the atmosphere by noxious impurities.

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* Part I can be found in Volume XII and Part II, in Volume XIII of the AICE Survey of USSR Air Pollution Literature.

** Izvestiya Vsesovuznogo Geograficheskogo Obschestva, Tom 101, vip. 4, Iyul' - Avgust 1969.

"On the first day, M. Ye Berlyand (Main Geophysical Observatory) reported on the status and prospects of development of meteorological studies of atmospheric pollution. He noted the following basic problems in the development of further research: study of meteorological conditions in the presence of heavy air pollution, including the mechanisms of turbulent exchange in the presence of inversions, a complex topography, and urban buildings; development of automatic and recording apparatus; study of the vertical distribution of noxious impurities; accumulation of observational data and development of methods of forecasting pollution, etc.

"In the report of I. L. Varshavskiy (State Committee of the USSR Council of Ministers on Science and Technology), 'Basic Trends of Scientific Research on Protection of the Air Reservoir from Pollution', the problems mentioned as the major ones involved the study of the biological action and hygienic importance of atmospheric pollutants, the influence of meteorological conditions on the distribution of noxious impurities in the atmosphere, the removal of noxious impurities from waste gases of industrial enterprises, and the removal of toxic components from the exhaust gases of internal combustion engines.

"Considerable interest was elicited by the report of M. S. Gol'dberg (A. I. Sysin Institute of General and Communal Hygiene) on the 'Hygienic Standards for the Maximum Permissible Content of Noxious Substances in Atmospheric Air.' He noted that hygienic standards worked out on the basis of experiments and actual laboratory and clinical studies constitute the scientific basis of environmental improvement measures in the struggle with atmospheric pollution, and make it possible to evaluate the results from the standpoint of providing the optimum living conditions for the population.

"Papers by a number of members of the Main Geophysical Observatory presented studies dealing with the physical validation of the procedure for calculating the pollution of the atmosphere with emissions of industrial enterprises and steam power plants, an investigation of turbulence in the lower 500-meter layer, an experimental study of pollution in industrial areas on the territory of the Soviet Union, the establishment of the relationship between meteorological conditions and air pollution in urban areas, the development of automatic methods of determination of atmospheric pollutants, etc. There were also reports on studies conducted in collaboration with Moscow University (S. M. Gorlin) in which the influence of irregularities of the earth's surface on the characteristics of the air flow was investigated in wind tunnels in connection with the problem of impurity diffusion.

"Members of the Institute of Experimental Meteorology (USSR) presented a series of reports on the study of diffusion of impurities and characteristics of turbulence from a 300-meter meteorological mast in Obninsk. Individual reports were also delivered by members of the Institute of Applied Geophysics

(Ye. N. Teverovskiy), the Central Aerological Observatory (N. Z. Pinus), the F. F. Erisman Scientific Research Institute of Hygiene (R. S. Gil'denskiol'd and B. V. Rikhter), and others.

"Specialists from Czechoslovakia (A. I. Vesecky, head of delegation), the German Democratic Republic (W. Warmbt, head of delegation), Poland (W. R. Parczewski, head of delegation) introduced the participants of the Symposium to the broad range of experimental research on the problem of atmospheric pollution. D. Szepesi (Hungary) reported on studies of meteorological conditions of turbulent diffusion, and G. I. Diaconescu (Rumania), on studies of thermal stratification of the atmosphere in connection with the pollution problem.

"All the participants of the Symposium came to the conclusion that at the present stage of the struggle with atmospheric pollution, the methods of purification and the construction of high stacks should be combined for the purpose of effectively utilizing the dispersing capacity of the atmosphere.

"The need for an extensive introduction of meteorological investigation of industrial projects and for the forecasting of air pollution was noted. One of the primary problems noted was that of validating and standardizing the methods of calculation of atmospheric pollution, the meteorological and hygienic procedures in studying the chemical composition of the atmosphere, and the meteorological parameters determining this composition."

It is hoped that the papers presented in this volume as well as in the preceding two volumes will be conducive to a better appreciation of the meteorological-air pollution investigations conducted in the USSR and in a number of the Soviet-block countries. As the editor of this volume I wish to thank my co-workers in the Air Pollution Section of the Institute for their valuable assistance.

M. Y. Nuttonson

May 1972

ON THE REMOVAL OF IMPURITIES FROM THE ATMOSPHERE BY CLOUDS AND PRECIPITATION

Ye. S. Selezneva and O. P. Petrenchuk (USSR)

From Glavnoe Upravlenie Gidrometeorologicheskoy Sluzhby Pri Sovete Ministrov SSSR. (Chief Administration of the Hydrometeorological Service Under the Council of Ministers of the USSR.) "Meteorologisheskie Aspekty Zagryazneniya Atmosfery". (Meteorological Aspects of Air Pollution.) Sbornik dokladov na mezhdunarodnom simpoziume v Leningrade - Iyul' 1968 g. (Reports delivered at the International Symposium in Leningrad - July 1968.) Pod redaktsiey d-ra fiz.-mat. nauk M. E. Berlyanda. (Edited by Prof. M. E. Berlyand.) Gidrometeorolgicheskoe izdatel'stvo, Leningrad, p. 253-259, (1971). (Hydrometeorological Publishing House, Leningrad, (1971).)

The last 10-15 years have seen the development of research in the chemistry of atmospheric precipitation in many countries. Work done along these lines by the Stockholm Meteorological Institute is widely known [9]. An annual cycle of precipitation composition studies was carried out in the U.S.A. under the direction of Yunge [8]. During the period of the International Geophysical Year, considerable work was done in Czechoslovakia [10], in the German Democratic Republic [11], and in other European countries. Since that time, systematic studies of the chemical composition of atmospheric precipitation have also been conducted by the A. I. Voyeykov Main Geophysical Observatory with the use of a selective network of hydrometeorological stations for collecting the precipitation samples [1].

The results of these studies, which thus far have been fairly numerous, may be treated from different points of view, in particular, from the standpoint of evaluating the spreading of pollutants from industrial and natural sources.

The chemical composition and total concentration of dissolved and undissolved impurities in precipitation show the degree of pollution of the atmospheric layers through which it falls. An idea of the propagation of atmospheric pollutants was obtained from the geographical distribution of the total mineralization of precipitation.

Studies made at the Main Geophysical Observatory and the compiled maps of distribution of the main impurities in the precipitation showed that in areas distant from industrial plants, under the purest atmospheric conditions, the precipitation mineralization (i.e., the total ions) amounts to 10-15 mg/l; in areas of appreciable natural and industrial atmospheric pollution, the average precipitation mineralization increases to 30-40 mg/l, and sometimes to higher values. Particularly polluted is the precipitation in industrial cities and their environs, where the mineralization frequency exceeds 100 mg/l [6].

Depending on the mineralization and amount of precipitation, from 5 to 15 tons of substances per 1 km^2 fall on the earth's surface with the precipitation each year over the territory of the USSR, and 20-30 tons or more fall on cities. The annual amount of dry deposits in cities is much greater, but it is essential to note that precipitation removes finer aerosols and gaseous impurities from the atmosphere, in particular, sulfur dioxide, which readily dissolves in water.

Thus, atmospheric precipitation is a highly important factor in the process of self-purification of the atmosphere. An approximate global estimate of the effectiveness of this process may be given.

On the basis of the above-mentioned studies conducted in different countries, an average precipitation mineralization of 20-30 mg/l may be assumed for the globe (probably, closer to 20 mg/l). Each year, 5 x 10^{14} tons of water, i.e., about 10^{10} tons of dissolved salts fall on the earth's surface. For comparison, let us indicate that the world's industry consumes 2 x 10^9 tons of coal per year. As a result of the combustion of this amount of coal, 4 x 10^7 tons of sulfur dioxide or 1.3 x 10^7 tons of sulfur is discharged into the atmosphere. Precipitation annually removes 10^8-10^9 tons of sulfur from the atmosphere (if the average sulfur concentration in the precipitation is taken to be 2-3 mg/l). Comparison of these data suggests that on a global scale, the industrial contribution is small and barely amounts to 5% of the impurities supplied by natural sources, i.e., by the world ocean, the surface of dry land, and volcanoes.

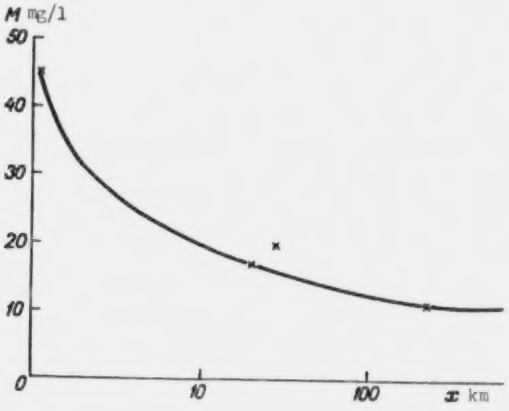

Fig. 1. Change of precipitation mineralization with distance from Leningrad to the east.

When the process is considered on different scales, the relationship is substantially altered. We will now switch from a global estimate to macro- and mesoscales.

As was noted above, in industrialized cities the precipitation mineralization increases markedly in comparison with rural areas. Fig. 1 shows the change of precipitation mineralization as one moves away from Leningrad. Within the confines of a large city such as Leningrad, the mean annual mineralization amounts to about 45 mg/l, and is smaller in summer and larger in winter. As one moves away from the city, the mineralization decreases rather quickly: at a distance of 10-15 km the total concentration of ions in the precipitation already decreases 2.5-fold, and at a distance of 100-150 km it changes into the background concentration of 10-11 mg/l, which is characteristic of this geographical zone. Such a rapid decrease of impurities in precipitation should be regarded as an indicator of intense scattering of the impurities in the atmosphere.

The chemical composition of precipitation begins to be formed in the cloud during the condensation of water vapor on condensation nuclei and absorption of various impurities by the cloud droplets. In order to evaluate the washing effect of precipitation, it is necessary to study all the stages of the process. It is necessary to have data on the composition of the water in the clouds, composition and concentration of atmospheric aerosols, and finally, composition of the precipitation. To obtain all these data, in addition to precipitation samples, cloud water and aerosol samples were taken from an airplane. Flights were carried out for this purpose over the four major regions of the European Territory of the USSR: north, northwest (mainly the Arkhangel'sk and Leningrad Oblasts), southwest (the Kiev-Dnepropetrovsk regions) and southeast (the Trans-Volga). In addition, a number of flights were made in Western Siberia (Krasnoyarsk Oblast).

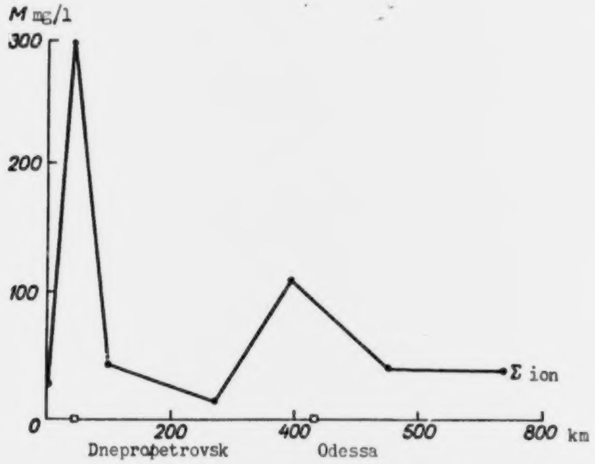

Fig. 2. Variation of cloud water mineralization in the horizontal direction during the flight of 7 December 1963.

From the standpoint of cloud water composition, precipitating clouds should be treated separately from internal stratified clouds. In all types of clouds,a large number of samples were collected which were analyzed in the laboratory under the direction of V. M. Drozdova.

Low stratified clouds, particularly those formed under inversions, accumulate impurities, which build up under the inversion and fully reflect the local contaminations of the lower atmospheric layer. Fig. 2 shows the results of a horizontal flight in such clouds along the Kiev-Dnepropetrovsk-Odessa route. The flight was conducted in the central portion of an intense anticyclone occupying the entire European part of the Soviet Union at a height of 500 m, under an inversion layer. Fig. 2 shows the sharp rise of the concentration of impurities in cloud water as one approaches the cities. Near Dnepropetrovsk, the cloud water mineralization reached 300 mg/l. In the absence of an inversion in this region, the mineralization of water in stratified clouds is 15 mg/l.

In the northern regions of the European Territory of the Soviet Union and in Western Siberia, the average mineralization of stratified clouds is 9-12 mg/l. However, under anticyclonic inversion conditions, the mineralization increases here as well, as may be seen in Fig. 3, which gives results of sounding in the area of Krasnoyarsk. The clouds from which the samples were collected were located at a comparatively high elevation (above 2000 m) under an anticyclonic inversion layer. However, the pollutants also reached up to this height. The total mineralization of cloud water exceeded 30 mg/l, i.e., reached a very high value for these regions, while the concentration of sulfur oxides (SO_4 ions) was 60-80%.

The chemical composition of the water from frontal precipitation clouds differs markedly from the stratified and stratocumulus clouds discussed. On the basis of an analysis of a large number of samples (around 90), the average mineralization of frontal clouds is 6 mg/l, and it is very stable in space and time.

Thus, in evaluating the contribution of clouds to the mineralization of precipitation, the initial value may be taken as 6 mg/l. Higher values of this quantity in precipitation are obviously caused by washing of the impurities out of the subcloud layer.

The process of washing of impurities out of the atmosphere is given considerable attention in the study of radioactive contaminants [3, 4, 7]. In theoretical discussions it is usually assumed that the rate of washout of impurities by precipitation is proportional to the concentration of impurities in the atmosphere $\frac{dq}{dt} = -\sigma q.$ Accordingly, the concentration of impurities in the atmosphere is expressed by the formula

$$q_t = q_0 e^{-\sigma t}, \qquad (1)$$

where σ is the washout coefficient.

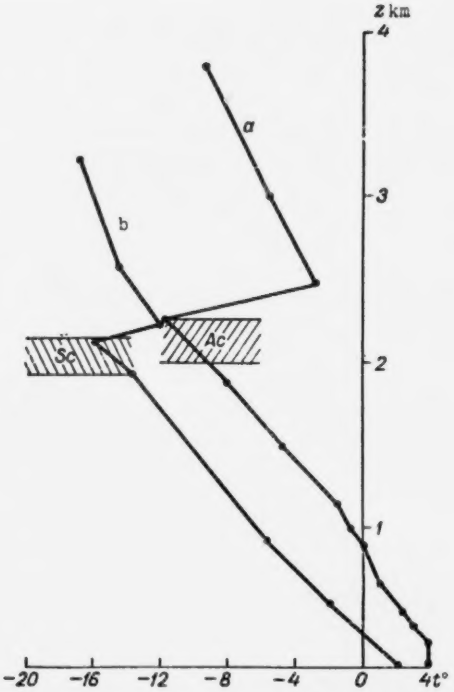

Fig. 3. Results of temperature sounding of the atmosphere in Krasnoyarsk.
(a) 19 December 1965, Σ_{ion} = 32 mg/l;
(b) 16 May 1966, Σ_{ion} = 31 mg/l.

The literature contains estimates of the coefficient σ, summarized in a review article by K. P. Makhon'ko [4]. For radioactive contaminants, $\sigma = 10^{-5}$ sec^{-1} is taken. For nonradioactive impurities, the available data on the determination of σ are very scarce. Here we can also refer to the work of Makhon'ko [4], who obtained the value 6.7×10^{-5} sec^{-1} for the washout coefficient of atmospheric dust, and the study of B. G. Andreyev, according to which σ is equal to 1.3×10^{-4} sec^{-1}. In the calculations below, the washout coefficient will be assumed equal to 10^{-4} sec^{-1}.

The amount of substance washed out of an atmospheric column of height H and cross sectional area s during time t = T may be expressed by the formula

$$\Delta Q = Hs \int_{q_{t=0}}^{q_{t=T}} dq = -Hsq_0\left(1 - e^{-\sigma T}\right). \qquad (2)$$

We then obtain the following formula for the concentration of impurities in precipitation:

$$M = M_{\text{обл}} + \frac{H}{h} q_0\left(1 - e^{-\sigma T}\right) \text{ мг/л}. \qquad (3)$$

where M_{cl} is the concentration of impurities in cloud water, equal to 6 mg/l, and h is the height of precipitation in mm.

Formula (3) was used to calculate the concentration of impurities in precipitation, certain assumptions being made. Thus, in particular, the height of the subcloud layer H was taken as 1000 m. On the basis of climatological studies [2], the average rate of rainfall was taken as follows: 0.6 mm/hr for region I, 1.0 mm/hr for II, 1.5 mm/hr for III, and 1.0 mm/hr for IV; the average duration of rainfall for the same regions was 5.6, 4.4, 3 and 3.4 hours respectively.

The calculation was made by taking into account P. F. Svistov's experimental data on the content and chemical composition of aerosols in the same regions in the 250-1000 m atmospheric layer. Maximum contents of the impurities were used, since these values give a better description of the degree of atmospheric contamination than average concentrations obtained on the basis of measurements both before and after precipitation. The maximum impurity contents in regions I, II, III and IV were 20, 60, 145 and 100 μg/m^3, respectively.

The contribution of the 0-250 m layer was estimated on the basis of a formula characterizing the vertical distribution of aerosol particles N [5]

$$N(z) = N_0 \frac{C^2}{(C+z)^2}, \qquad (4)$$

where N_0 is the concentration of aerosol particles near the ground, and C is some integration constant equal to about 10^5 cm, according to experimental data for average conditions.

It follows from formula (4) that the content of aerosol particles in the atmospheric column is

$$\int_0^z N(z)\,dz = N_0 \frac{C^2}{C+z}\Big|_z^0, \qquad (5)$$

and hence, the 0-1000 m layer contains 50% of all aerosols, and the 0-250 m layer, 20%.

Results of the calculations made with formula (3) are given in Tables 1 and 2.

Table 1 shows what fraction of precipitation impurities is determined by the contribution of cloud water and what fraction is washed out by precipitation in the subcloud layer. In the northern regions under pure atmospheric conditions, the chief contribution (over 50%) is made by clouds, i.e., largely condensation nuclei of marine origin. In southern regions with heavy atmospheric pollution, the predominant process is washing out, and 70-80% of all the impurities are acquired in the subcloud layer.

Table 1

Amount of Impurities (%) Acquired by Precipitation from Clouds and from the Subcloud Layer

	Region			
	I	II	III	IV
Clouds	55	35	22	23
Subcloud layer	45	65	78	77

Table 2

Mineralization of Atmospheric Precipitation (mg/l)

Method by Which Obtained	Region			
	I	II	III	IV
Calculated from formula (3)	10,9	16,9	27,0	26,3
Actual average for several years	14,1	16,4	30,5	24,8

In Table 2, the precipitation mineralization calculated from formula (3) is compared with the actual mineralization. A satisfactory agreement of the data confirms that the adopted washout coefficient, 10^{-4} sec^{-1}, may be considered characteristic of nonradioactive aerosols.

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E. S. SELEZNEVA, O. P. PETRENCHUK

ON THE REMOVAL OF POLLUTANTS FROM THE ATMOSPHERE BY CLOUDS AND PRECIPITATION *

An important factor of atmosphere selfcleaning is atmospheric precipitation, the chemical composition of which is determined by two processes: by water vapour condensation on condensation nuclei, and by catching pollutants both in cloud and subcloud layers with cloud and precipitation elements.

The chemical composition and total concentration of soluble and insoluble pollutants in precipitation is pollution index of those atmospheric layers through which they fell out. Precipitation mineralization and sulphur oxide content in it increase substantially in industrial towns and regions.

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* Editor's note: The abstract is presented as given in English with the original Russian article.

Clouds contribute, to some extent, to chemical composition of precipitation. Investigations carried out in the Main Geophysical Observatory show that the chemical composition of cloud water depends greatly on meteorological factors. Subinversion clouds formed under anticyclonic situation are peculiar "accumulators" of pollutions concentrated in subinversion layer.

The chemical composition of water from frontal clouds which produce precipitation is nearly identical for different regions and changes little with time during precipitation fallout. Mineralization of cloud water from frontal clouds is equal to 6 mg/litre.

The contribution of cloud and subcloud layers to formation of chemical precipitation composition is determined on the basis of experimental data and calculations. The evaluation shows that in the regions with weak pollution the main amount of pollutants (more than 50%) comes from clouds, and in the regions with heavy air pollution washing-out becomes the prevailing process: 70—80% of all pollutants are captured in subcloud layer.

SOME ASPECTS OF THE ADOPTION OF AUTOMATIC METHODS OF DETERMINING ATMOSPHERIC POLLUTANTS

N. Sh. Vol'berg, G. V. Gal'dinov, and V. Z. Al'perin (USSR)

From Glavnoe Upravlenie Gidrometeorologicheskoy Sluzhby Pri Sovete Ministrov SSSR. (Chief Administration of the Hydrometeorological Service Under the Council of Ministers of the USSR.) "Meteorologisheskie Aspekty Zagryazneniya Atmosfery". (Meteorological Aspects of Air Pollution.) Sbornik dokladov na mezhdunarodnom simpoziume v Leningrade - Iyul' 1968 g. (Reports delivered at the International Symposium in Leningrad - July 1968.) Pod redaktsiey d-ra fiz.-mat. nauk M. E. Berlyanda. (Edited by Prof. M. E. Berlyand.) Gidrometeorolgicheskoe izdatel'stvo, Leningrad, p. 260-268 (1971). (Hydrometeorological Publishing House, Leningrad, (1971).)

At the present time, a number of instruments are being designed in the Soviet Union for the continuous determination of the basic atmospheric pollutants. Their adoption will make it possible to increase substantially the amount of information obtained, eliminate its delay, and create automatic systems to warn of the possibility that the maximum air pollution norms may be exceeded.

The expedience of reducing as much as possible the number of available gas analyzer models lends great urgency to the problem of selecting the most rational principles of determination of the substances in question.

Instruments designed for analysis of atmospheric air should be characterized by a high sensitivity. Their transportability is also important. We shall discuss the most important types of existing gas analyzers from these points of view.

The highest sensitivity is that of ionization instruments. Thus, flame ionization gas analyzers used for the determination of total hydrocarbons in air permit the determination of thousandths of mg/m^3, have a short time lag (of the order of a few seconds), and may be constructed in portable form. For example, the portable flame-ionization gas analyzer designed by Guerrant [8] has a sensitivity of the order of 10 $\mu g/m^3$ and a time lag of 2 sec. The instrument is completely self-contained and weighs 12 kg.

For the determination of a large number of inorganic gases, analyzers have been proposed that are based on the conversion of the component being determined to the aerosol state by the addition of a suitable gaseous or vaporous reagent to the air stream. The aerosol formed is determined from the decrease of the ionization current. The "Billion Air" American gas analyzer [5] and the domestic "Sigma-1" analyzer,* both operating on this principle, insure a high sensitivity of the determinations, but are stationary. There is reason to believe that portable instruments of this type will appear in the near future.

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* Automatic continuous-action "Sigma-1" gas analyzers. Prospectus of VDN Kh (Exhibition of Achievements of the National Economy of the USSR), 1967.

Among ionization gas analyzers, those whose specificity stands out are mass spectrometric instruments, particularly when they are combined with chromatographs. However, the complexity and high cost of the required equipment and the need of constant and highly qualified operation make their wide application to the determination of the main atmospheric pollutants impractical (at least in the next few years). Moreover, these instruments are not interchangeable in many types of research work.

To determine the qualitative and quantitative composition of atmospheric pollutants of organic character, unquestionably the most promising instruments are chromatographic analyzers equipped with various ionization detectors. Thus, the design of specialized portable or transportable instruments of this type is highly desirable.

Among instruments based on physical principles we should mention gas analyzers utilizing the absorption of light in the infrared region of the spectrum. These instruments are widely used for the determination of CO, CO_2 and CH_4 in various gases. Their advantage is a high specificity, a short time lag and simplicity of operation. However, the sensitivity of most of them is insufficient for the determination of CO in atmospheric air. The sensitivity of these instruments is usually raised by increasing the cell length. Thus, in the American gas analyzer "Lyra," designed for the determination of CO in atmospheric air, the cell size is 1 m. Obviously, these instruments can only be stationary. In the last few years, a certain progress has been made in regard to the reduction of their weight and size through the use of multipath cells or an open beam.

Photocolorimetric methods, both manual and automatic, are most widely used at the present time for the determination of air pollution. Two types of automatic photocolorimetric gas analyzers are used, liquid and belt types (Table 1).

In the principle of their operation, photocolorimetric gas analyzers are versatile instruments permitting the determination of any substance for which a rapid and sufficiently sensitive color reaction exists. This makes them convenient for incorporation into sets consisting of several instruments permitting the simultaneous determination of a number of substances [11].

A typical representative of modern liquid photocolorimetric gas analyzers for determining atmospheric pollutants is the "Imcometer" instrument [7]. It is designed for determining ozone, sulfur dioxide, nitrogen oxides, hydrogen sulfide, chlorine, and fluoride ions.

The working cycle of this instrument includes a series of successive operations: measuring out the reaction liquid, automatic setting of the null point, passing of the air under analysis, measurement, and emptying of the cell. The performance of all these switching operations requires suitable automatic devices, and this accounts for the comparative complexity of this type of instruments.

Table 1

Photocolorimetric Gas Analyzers

Instrument	Component determined	Measurement range, mg/m³	Basic error, %	Reference
Liquid type				
"Chromoflux"	Cl_2	0—20	10	Catalog of West German firm"FRG"
FK-1501	NO_2	0—5	10	[4]
"Imcometer"	O_3	0—0.2	5	[7]
Belt type				
FL-5501	NO_2	0—1	10	[4]
FGTs-ID	H_2S	0—0.1	20	[3]

Gas analyzers in which the reagent is applied on a paper or cloth belt have considerable advantages in this respect. These instruments can be made transportable much more simply and easily. Their major drawback, however, is a comparatively low accuracy, which usually amounts to 20%.

In order to obtain sufficient sensitivity in measurements with photocolorimetric instruments, the substance being determined should be collected from a comparatively large volume of air, 10 ℓ or larger. This accounts for the substantial lag of photocolorimetric gas analyzers, which in many cases exceeds 30 minutes.

A high sensitivity is easiest to obtain in instruments based on the use of electrochemical methods. The most widely employed of such gas analyzers have devices based on the conductometric principle, consisting in the measurement of the conductance of the reagent solution after reaction with the gas being analyzed. A brief description of the best known conductometric gas analyzer is given in Table 2.

The conductance of electrolytes strongly depends on the temperature, and all conductometric gas analyzers are therefore provided with thermostats. This is given particular attention in instruments designed for determining traces. Thus, in one of the comparatively new and most advanced instruments of this type, the "Picoflux" gas analyzers, all the heat-sensitive elements are placed in a precision liquid thermostat with thick plexiglas walls [6]. The thermostat is provided with a film heating element and a special centrifugal stirrer that make it possible to maintain the specified temperature of 40° C. to within 0.002°.

Table 2

Conductometric Gas Analyzers

Instrument	Components determined	Measurement limits for mg/m^3	Reference
"Ul'tragaz-3	SO_2, H_2S, NH_3, CO	0–5	Catalog of West German firm "FRG"
"Ionoflux"	SO_2, CS_2, PH_3 и др.	0–5	Same
"Picoflux"	SO_2, HCl, NH_3	0–1	[6]
"KU-3"	CO, Gasoline vapors	0–50 (CO)	[2]

The need for precise thermostating leads to an increase in the size and weight of the instrument. Thus, the "Picoflux" gas analyzer, which is considered to be transportable, weighs about 35 kg without the recorder.

Among electrochemical gas analyzers, the most interesting are instruments based on the coulonometric principle. This principle has been used in electrochemistry for a long time and on a wide scale, but its special advantages in gas trace analysis were revealed only in the last few years thanks to the work of Hersch [9], Novak [10], and several other investigators.

Coulonometric gas analysis is based on the measurement of the current of the electrode reaction entered into by the substance being analyzed, which is continually supplied to the coulonometric cell with the stream of the analyzed gas. In many cases, the reaction proceeds with a 100% current efficiency based on the substance. This makes it possible to use Farraday's law to calculate the concentration being measured without first having to calibrate the instrument. Participation in the reaction of all the substances supplied makes for a high sensitivity of the measurements, and this sensitivity can be achieved without using current amplifiers.

In contrast to other electrochemical methods (for example, conductometric and polarographic), the magnitude of the current is determined in this case only by the amount of electrochemically active substance entering the cell, and is independent of temperature. This makes it unnecessary to thermostat the instrument accurately and promotes a compact design.

In the USSR, the Experimental Design Office for Automation (OKBA) has developed and is currently introducing on a large scale a coulonometric gas analyzer GKP-1 for determining sulfur dioxide in atmospheric air [1]. The instrument makes use of the principle, proposed by Novak [10], of absorption of sulfur dioxide by a solution of iodine in sulfuric acid followed by electrooxidation of the iodide ions formed.

In contrast to the instruments proposed earlier, the GKP-1 instrument uses a nonflow-type arrangement, i. e., the electrolyte in the cell is not changed. A slight loss of electrolyte, which is due to the bubbling air, is compensated from a small tank built into the cell by means of an original arrangement for maintaining a constant level.

Such a design could be realized thanks to the fact that in a coulonometric determination, in contrast to other electrometric and photometric methods, the reagent solution is continually regenerated.

Table 3 summarizes the chief technical characteristics of the most important gas analyzers, using the determination of sulfur dioxide as an illustration.

Table 3

Characteristics of the Most Important SO_2 Gas Analyzers

Principle of operation of gas analyzer	Brand	Scale, mg/m^3	Lag, min	Basic error, %	Weight, kg
Photocolorimetric	"Imcometer"	0–0.6	30	5	
Conductometric	"Picoflux"	0–1.0	1,5	6	35
Coulonometric	GKP-1	0–1.0	0,75	6	7

It is evident from the above data that the coulonometric gas analyzer has the most desirable set of properties. Having a sensitivity and accuracy like those of the best photocolorimetric and conductometric instruments, the coulonometric gas analyzer is distinguished by a short lag and small weight. The absence of moving parts and a long service life after charging make the instrument operationally reliable and one that requires a minimum of handling and of operating time.

The indicated advantages of coulonometric gas analyzers place them in the rank of the most promising instruments for determining atmopsheric pollutants, and make it expedient to expand as much as possible the number of substances for the determination of which they can be used. At the present time, instruments are being developed for the determination of hydrogen sulfide, chlorine, ozone, hydrogen chloride, etc.

The transportability of the above coulonometric gas analyzer enabled us to make a study of the atmospheric pollution of Leningrad both at ground level, with the instrument mounted on a truck, and at a height of 100 m, with the instrument carried in a helicopter. The results obtained are illustrated in Figs. 1 and 2.

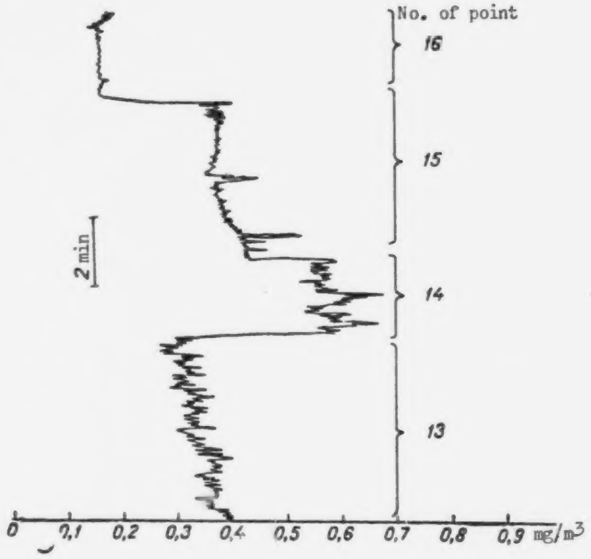

Fig. 1. Measurement of sulfur dioxide concentrations in the city by means of the GKP-1 instrument mounted on a truck.

Measuring time at one point, 3-5 min; number of points of measurement in the course of a working day, 25-30.

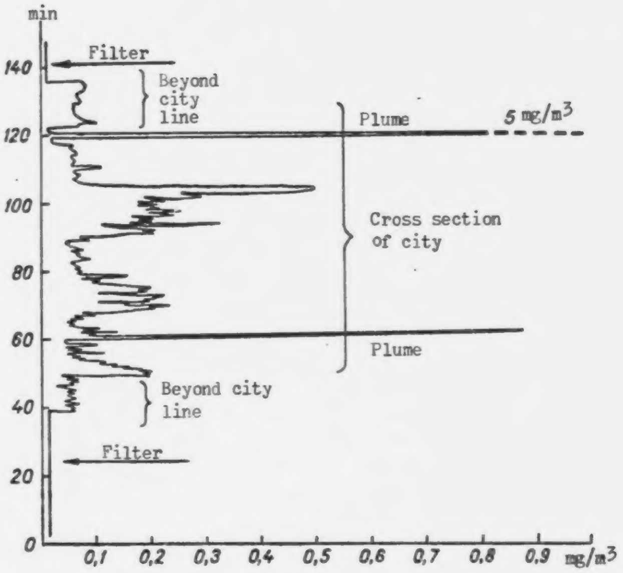

Fig. 2. Measurement of sulfur dioxide concentrations with GKP-1 instrument mounted on helicopter.

Flight route perpendicular to the wind direction; flight altitude 100 m; speed, 60-70 km/hr.

Thus, to determine the chief inorganic atmospheric pollutants, the most promising approach appears to be the development of instruments based on the coulonometric principle.

Because of the considerable increase in the volume of information that can be obtained when automatic instruments are used, the problems of processing of this information also assume a major importance. For example, the digitizing of tape carrying the recording of concentration variations with time frequently takes more time than the recording itself. Automation of this process, however, is an independent and fairly difficult problem. Moreover, without preliminary digitizing, a continuous recording does not provide the answers to the following questions that are usually of interest to hygienists and meteorologists: what is the average concentration for the time interval in question and what is the number of cases in which the maximum permissible concentration has been exceeded by a certain factor? For this reason, it is our view that in many cases it is possible and expedient to abandon the traditional use of recorders as secondary instruments and to replace them with integrating devices in combination with the simplest amplitude analyzers. In our view, the development of small and reliable instruments of this type must not be delayed.

When automatic gas analyzers are used for the determination of air pollutants, the problems of systematic checking of their readings assume a major importance. To provide for such control on the spot, it is necessary to develop simple and reliable measuring devices and also to create a center for preparing standard gas mixtures.

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N. Sh. VOLBERG, G. V. GALDINOV, V. Z. ALPERIN

ON THE AUTOMATIC TECHNIQUE FOR PROCESSING OF INFORMATION ON AIR POLLUTION *

At present, in the Soviet Union a series of instruments for continuous measurement of air pollution is developed. The necessity of minimizing the number of types of gas analyzers makes the problem of introduction of highly sensitive transportable instruments quite actual.

Ionization methods are especially highly sensitive, in particular, ionization method of hydrogen flame. A description of a transportable gas analyzer of this type destined for hydrocarbon determination in the air is given.

Among other ionization gas analyzers, mass-spectrometers are of special interest. But the complexity of these instruments makes it expedient to use them only for investigation purposes.

The most perspective instruments for quantitative and qualitative analysis of air pollution are chromatographic analyzers supplied by different ionization detectors. Therefore, the development of special transportable instruments of this kind is very desirable.

For carbon oxide determination, the optic-acoustic principle is rather perspective. The instruments of this kind are developed which possess high sensitivity. For instance, gas analyzer *GMK-3* has a scale of 0—40 mg/m^3 and is transportable.

Widely used at present are liquid photocolorimetric gas analyzers, but they have rather complicated mechanism and large sizes. Necessary sensitivity is usually reached by concentration of determined substance from large air volume; this stipulates substantial inertia of these gas analyzers.

Instruments based on electrochemical principles are more sensitive. Among them, coulonometric analyzers possess especially favourable features and high sensitivity along with small weight and sizes. So, developed in the USSR, transportable coulonopolarographic analyzer on sulphur dioxide *GKP-1* has sensitivity and precision of the best photocolorimetric and conductometric instruments. High metrological qualities of coulonometric gas analyzers place them among the most perspective instruments for air pollution determination and make it expedient to enlarge the number of substances determined by them.

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*Editor's note: The abstract is presented as given in English with the original Russian article.

To simplify the treatment of large information volume received from automatic instruments, it is expedient to refuse, in some cases, the utilization of recorders at the exit, employing integrating mechanisms instead of them coupled with the simplest amplitude analyzers.

Systematic control of automatic instruments is one of the most important conditions for receiving reliable measurement results. Development of precise dosimeters is necessary for supplying the possibility of conducting *in situ* control.

RECORDING OF SULFUR DIOXIDE CONTENT AT THE OUTSKIRTS OF A CITY. COMPARISON OF MEASUREMENT RESULTS FOR A VALLEY AND AN ELEVATION

H. Mrose and W. Warmbt GDR (German Democratic Republic)

From Glavnoe Upravlenie Gidrometeorologicheskoy Sluzhby Pri Sovete Ministrov SSSR. (Chief Administration of the Hydrometeorological Service Under the Council of Ministers of the USSR.) "Meteorologisheskie Aspekty Zagryazneniya Atmosfery". (Meteorological Aspects of Air Pollution.) Sbornik dokladov na mezhdunarodnom simpoziume v Leningrade - Iyul' 1968 g. (Reports delivered at the International Symposium in Leningrad - July 1968.) Pod redaktsiey d-ra fiz.-mat. nauk M. E. Berlyande. (Edited by Prof. M. E. Berlyand.) Gidrometeorolgicheskoe izdatel'stvo, Leningrad, p. 269-280, (1971). (Hydrometeorological Publishing House, Leningrad, (1971).)

1. Formulation of the Problem

In characterizing the air pollution in a city, much attention is given to the distribution of sulfur dioxide over the city and its environs. At the same time, it is assumed that the sulfur dioxide concentration in densely populated areas is markedly different because of the presence of a large number of different emission sources. In order to find out how representative the results of a single point of measurement are for a large area, we recorded the sulfur dioxide concentration at two points on the outskirts of Dresden from 12 January to 14 April 1968. The measurements involved the use of two sulfur dioxide recorders of Novak's design, kindly supplied to us for testing by the Hydrometeorological Service of CSSR.

2. Location of the Points of Measurement

The point of measurement at Wahnsdorf was located near the Wahnsdorf Meteorological Observatory at an elevation of 246 m above sea level and about 100 m above the valley.

The point of measurement at Radebeul was located in the Meteorological Service building at an elevation of 122 m above sea level. The Radebeul region consists of a residential area surrounded by gardens and green plantings. The distance between the points was 1.4 km. There were no major industrial plants around these points within a distance of approximately 3 km.

3. Method of Measurement of Sulfur Dioxide Concentration after Novak

The method of measurement of sulfur dioxide concentration after Novak [5, 6] is based on the following principle. Air is drawn through sulfuric acid containing iodine. In this process, sulfur dioxide reduces an equivalent amount of iodine to iodide ions according to the reaction

$$SO_2 + I_2 + 2H_2O = SO_4^{--} + 4H^+ + 2I^-.$$

The iodide ions formed are reoxidized to iodine on a platinum anode. The electric current required by this process is a measure of the iodide ions and thus of the sulfur dioxide concentration. Use of Faraday's law permits the calculation of the amount of iodine ions and hence, of the amount of sulfur dioxide from the measured current intensity. Knowing the equivalent weight of sulfur dioxide, Faraday's constant, and the suction rate of air, one can determine the sulfur dioxide concentration from the formula

$$SO_2 = \frac{21{,}1}{v} i \ (\mathrm{mg/m^3}),$$

where i is the measured current (μA) and v is the flow rate of air (ml/min).

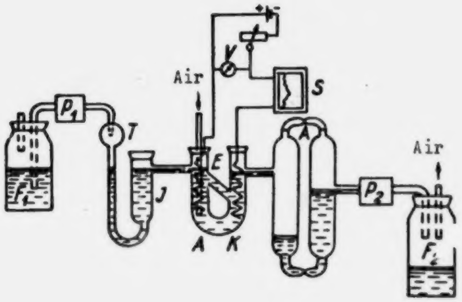

Fig. 1. Novak's gas analysis apparatus for determining sulfur dioxide.

E - electrolysis vessel, A - leveling vessel, P_1 - pump; T - attachment for drop formation, J - vessel for iodine, F_1 - acid, F_2 - acid used, P_2 - suction pump, S - recorder, V - voltmeter.

A diaphragm pump was used to introduce a 3% sulfuric acid solution into the anode compartment through a glass vessel filled with iodine crystals (Fig. 1). The air studied was also passed through this vessel. This air together with the sulfuric acid solution then entered the cathode compartment, from which it was drawn together with the spent sulfuric acid by a teflon pump. A voltage of about 0.3 V was applied to the electrodes. This voltage was less than required for decomposition of sulfuric acid, so that the current was maintained only by the oxidation of iodine ions. The measured current was recorded with a recorder having a scale from 0 to 50 mA and a clutch mechanism. The chart speed was 2 cm/hr.

4. Testing of Novak's Method and of the Measuring Instrument

The readings of Novak's analyzer were checked by comparing them with sulfur dioxide concentration measurements obtained by the specific method of West and Gaeke [8], which involves the use of para-rosaniline and sodium tetrachloromercurate (TCM). We obtained the following equation for regression curve, relating the data of Novak's method (y) to those of the TCM method (x):

$$y = 1{,}050x + 0{,}002 \ (\mathrm{mg/m^3})$$

with a confidence interval (P = 95%) for the constants

$$\Delta b = 0{,}182, \quad \Delta a = 0{,}035.$$

Comparative measurements made by Herrmann [3] in the industrial district of northern Bohemia led to nearly identical results (Table 1). It follows from the table that both methods yield practically the same results.

Table 1

Comparative Measurements of Sulfur Dioxide Concentration Made by Using Novak's Method and the TCM Method

	r	a	b
For Wahnsdorf	0,97	0,002	1,05
After Herrmann	0,96	−0,001	0,99

Note. r is the correlation coefficient.

In order to check the reproducibility of the data obtained by Novak's method, we conducted parallel measurements at Wahnsdorf by using two of Novak's instruments. We thus obtained the following regression equation for the readings of the first instrument (x) and second instrument (y)

$$y = 0{,}961x - 0{,}003 \text{ (mg/m}^3\text{)}$$

with confidence intervals (P = 95%) for the constants

$$\Delta b = 0{,}033, \quad \Delta a = 0{,}005.$$

The confidence interval for the standard deviation of the individual quantities from the regression curves for P = 95% is

$$\Delta S = 0{,}03 \text{ (mg/m}^3\text{)}.$$

The inaccuracy of the readings is thus equal to approximately one part per million (10^{-6}).

5. Evaluation of Measurement Results

The half-hourly averages for the first 30 minutes were determined by graphical interpolation of the data of readings given by Novak's instrument.

Table 2 lists the arithmetic averages D for all the measurement results, and also for 0, 6, 12 and 18 hours. It is characteristic that only slight differences in concentration exist between Wahnsdorf and Radebeul. An examination of the average values shows that the sulfur dioxide content in the valley (Radebeul) is approximately 20% higher than on the elevation (Wahnsdorf). This difference is explained, on the one hand, by the fact that industrial plants and residential areas are concentrated in the valley, and on the other hand, by the fact that the conditions produced in the valley are not as conducive to the dispersal of emissions as the conditions associated with the elevation, particularly in stagnant situations.

Table 2

Average Values of Sulfur Dioxide Measured by Novak's Method in Wahnsdorf from 12 January to 14 April 1968

Point	General Average		0 Hours		6 Hours		12 Hours		18 Hours	
	n	*D*	*n*	*D*	*n*	*D*	*n*	*D*	*n*	*D*
Radebeul	2172	0,117	270	0,085	272	0,136	271	0,143	271	0,103
Wahnsdorf	2085	0,099	264	0,084	260	0,121	263	0,121	265	0,084
Difference		0,018		0,001		0,015		0,022		0,019

Note. n - number of measurement; calculation of average values for fixed hours also included data of observations made one hour before and one hour after the observations.

The average daily variation of sulfur dioxide concentration in Radebeul and Wahnsdorf shows a difference in amplitude but not in phase (Fig. 2). The experimental values at both points were observed at the same time; the maximum concentration appeared around 9 A.M., and the minimum concentration, around 2 P.M. The average value of the daily fluctuation was 0.09 mg/m^3 in Radebeul and 0.07 mg/m^3 in Wahnsdorf. The maximum in the early morning hours is explained by the start of operation of industrial plants and a negligible turbulent exchange of the air masses in the presence of a stable atmospheric stratification. Heating in the evening hours causes only a slight increase in concentration, which is weakly manifested at the points of measurement around 6 and 7 P.M. The concentration minimum around 2 A.M. is explained by a slight emission during the nighttime hours. It is interesting to note that because of a better exchange, the sulfur dioxide concentration at the elevation changes little during the period from 4 P.M. to 2 A.M.

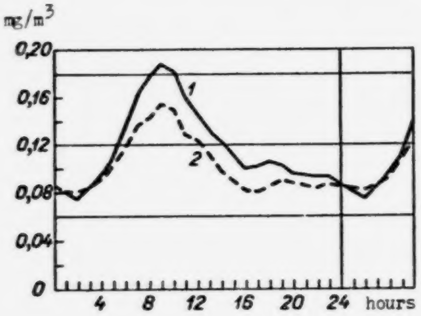

Fig. 2. Average daily variation of sulfur dioxide concentration in Radebeul (1) and Wahnsdorf (2).

To get an idea of the recurrence of any given sulfur dioxide concentrations, Figs. 3 and 4 show the distribution of total frequencies for the periods of 0, 6, 12, and 18 hours. Since the sulfur dioxide concentration values are distributed in accordance with the lognormal law, not the normal

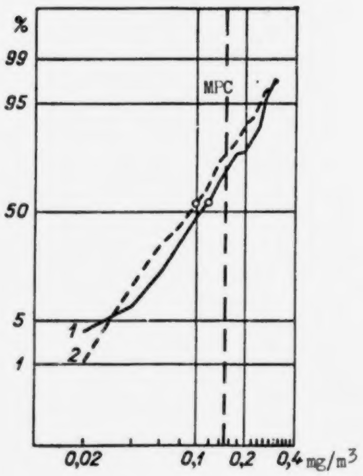

Fig. 3. Distribution of total frequency of average daily SO_2 values in Radebeul (1) and Wahnsdorf (2).

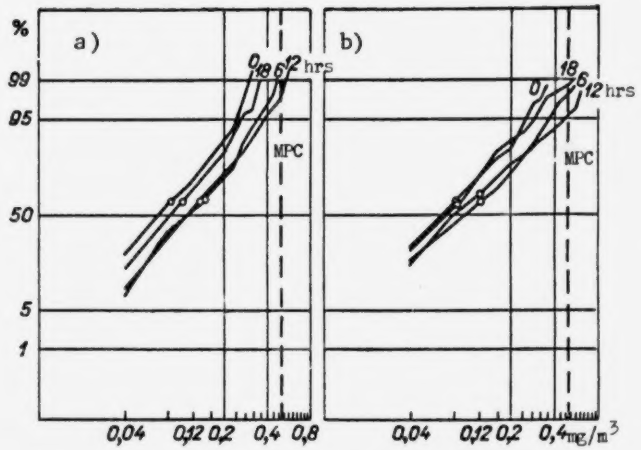

Fig. 4. Distribution of total frequency of average concentration values for different observation periods in Radebeul (a) and Wahnsdorf (b).

law, they are arranged along a straight line on a logarithmic probability plot. No large differences are observed between Radebeul and Wahnsdorf. It also follows from Table 3 that the total concentration values with 95% frequency, which were used in the literature in different ways to determine pollution, differ insignificantly for the two points. It should be noted that concentration values with 95% frequency in Wahnsdorf substantially surpass the corresponding values in Radebeul at 12 M. and less substantially at 6 A.M. and 6 P.M. It follows that the sulfur dioxide concentrations at the elevation are sometimes higher than in the valley. We are referring to cases in which emissions from industrial plants and residential sections in Dresden-Radebeul are carried by southeastern winds in the direction of the elevation.

Table 3

Total Sulfur Dioxide Concentration (mg/m^3) with 95% Frequency

Point	Average Value For Period	Time, Hours			
		0	6	12	18
Radebeul	0,25	0,25	0,35	0,39	0,27
Wahnsdorf	0,24	0,25	0.37	0,43	0,29

In order to establish the relationship between the average daily values of the two series of measurements, correlation coefficients were determined and the regression curve was plotted. The calculations were not made by using all the average daily values, but only values for the 1st, 4th, 7th days, etc., since consecutive mean daily values are not independent from one another. The correlation coefficient between the measured values in Wahnsdorf (x) and Radebeul (y) is

$$r = 0.918$$

The regression curve is expressed by the equation

$$y = 1{,}154x + 0{,}005 \ (mg/m^3).$$

For P-95%, the constants have confidence intervals

$$\Delta b = 0{,}192, \quad \Delta a = 0{,}018.$$

The results confirm the already-stated slight differences in sulfur dioxide concentrations between the valley and the elevation.

Let us consider in more detail some individual examples of recording of like and unlike daily variation of sulfur dioxide concentrations at the two points.

The variation of half-hourly average concentration values for the period of 4-10 February 1968 shows a close relationship between these values at Radebeul and Wahnsdorf (Fig. 5).

As an example of like variations of sulfur dioxide concentrations, let us consider the tracing of the recording of 27 January (Fig. 6). In the valley and at the elevation, an increase of the sulfur dioxide concentration was observed at about 12 M., and a decrease was noted past 2 P.M. From the wind velocity and direction tracings in Wahnsdorf one cannot infer the possible causes of these fluctuations. It is possible that the gaseous emissions were spreading through the valley in a broad front. Such changes are by no means explained, as is frequently done, by local influences such as emissions from neighboring houses.

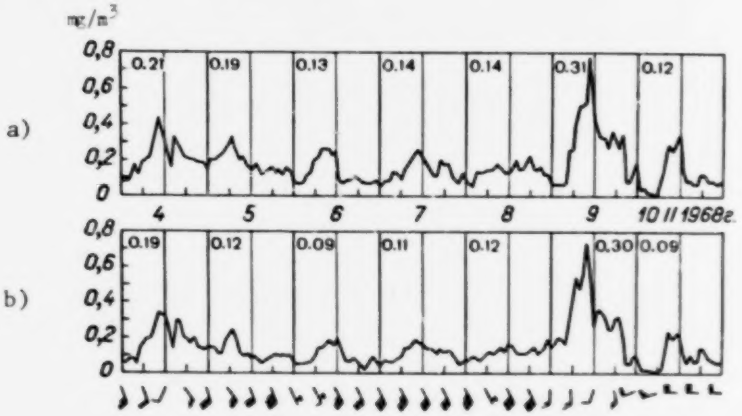

Fig. 5. Variation of sulfur dioxide concentration (average values obtained) in Radebeul (a) and Wahnsdorf (b) from 4 to 10 February 1968.

An unlike variation of sulfur dioxide concentration is illustrated by observations of 14 April 1968 (Fig. 7). On that day, appreciable concentration fluctuations were observed in Wahnsdorf, whereas they were absent in Radebeul. An explanation of this may be found by examining the recording of wind gustiness in Wahnsdorf. In the southeastern flow, short-term fluctuations in wind direction and velocity were observed since midnight, during which the sulfur dioxide concentration decreased as a result of turbulent mixing. One is struck by the brief increase in sulfur dioxide concentration at about 8 A.M. in Wahnsdorf. Concentration peaks of this kind are observed when the earth's surface is heated and the night surface inversion breaks down as a result of thermal convection. At the same time, the sulfur dioxide accumulated under the inversion spread toward the ground as a result of suddenly produced turbulent motions. In Radebeul, the inversion broke up later than at the elevation, i.e., at about 9 A.M.

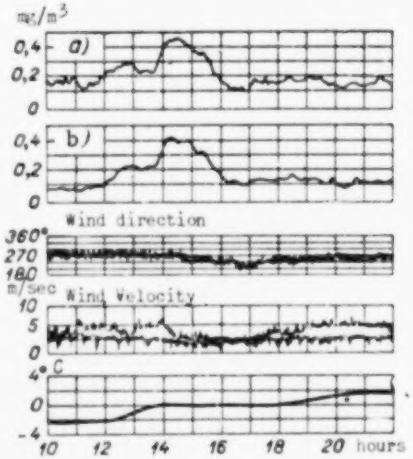

Fig. 6. Like variation of sulfur dioxide concentration in Radebeul (a) and Wahnsdorf (b), 27 January 1968.

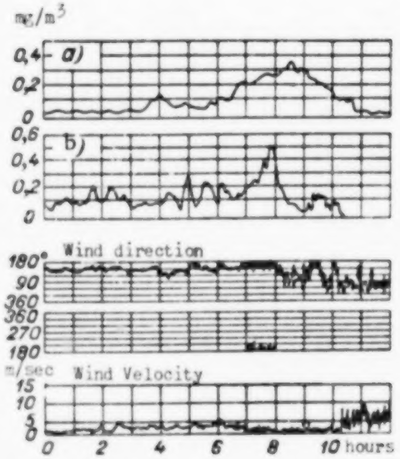

Fig. 7. Unlike variation of sulfur dioxide concentration in Radebeul (a) and Wahnsdorf (b), 14 April 1968.

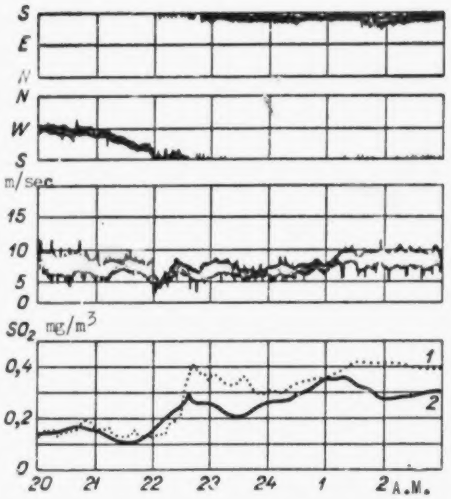

Fig. 8. Variation of sulfur dioxide concentration with changing wind direction in Radebeul (1) and Wahnsdorf (2) at -11 and -15°, 13-14 January 1968.

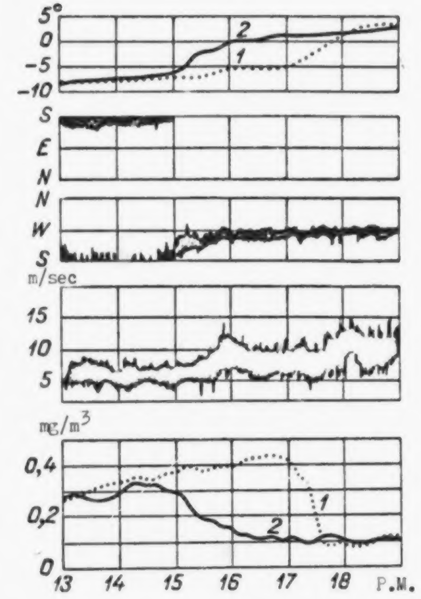

Fig. 9. Variation of sulfur dioxide concentration during passage of warm front.
1 - Radebeul, 2 - Wahnsdorf.

After the breakup of the inversion, fluctuations of wind direction caused by convection were observed at the elevation. The sulfur dioxide concentration decreased rapidly, since the wind changed direction from south-south-east to east, and no more air polluted with smoke emissions reached the point of measurement. An intensification of the wind after 10 A.M. led to a further mixing and thus to a decrease in sulfur dioxide concentration. It is evident from this example that a difference in sulfur dioxide concentration in the valley and on the elevation appears when local circulation systems arise under conditions of attenuated exchange.

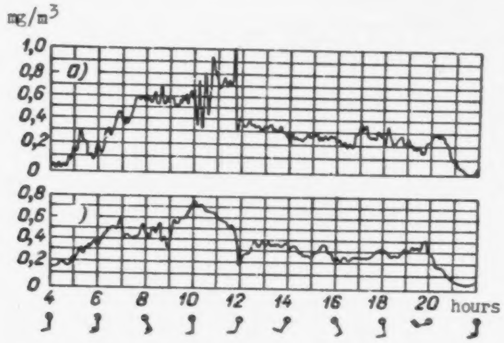

Fig. 10. Variation of sulfur dioxide concentration, 9 February 1968.

In addition, differences in sulfur dioxide concentration were observed in the presence of unlike wind direction and velocity and unlike turbulent state in the valley and on the elevation, as for example on 14 January 1968 during a clearly manifested passage of a warm front. On 13 January, Central Europe was under the influence of a cyclone located over Iceland. At about 10:30 P.M., the wind direction in Wahnsdorf changed to south-south-east (Fig. 8), and air with a high sulfur dioxide content began spreading from Dresden to both points. Warm air coming from the west reached Wahnsdorf in the afternoon hours on 14 January (Fig. 9). The passage of the warm front was confirmed by temperature and wind recordings (15^{20}). As the wind intensified and turned in a west-south-west direction, the mixing increased, and the concentration decreased. At that time, Radebeul was still surrounded by cold air. While the sulfur dioxide concentration on the elevation had already decreased, it was still increasing in the valley under a quasi-horizontal interface acting as barrier layer. Around 5 P.M., the warm air reached the valley, and sulfur dioxide concentration decreased rapidly.

In conclusion, we should also consider the sulfur dioxide variation of 9 February 1968. On that day, the highest concentrations of the 1967-68 winter season were measured (Fig. 10), and the valley of the Elbe was shrouded in mist and fog with weak southern winds.

As a result of small-scale circulation processes, large fluctuations of sulfur dioxide concentration were observed in the morning hours at both points. Later, because of an absence of exchange, the concentration in the valley and on the elevation increased. In Wahnsdorf, the maximum concentration was 0.75 mg/m^3 (10 A.M.), and in Radebeul it reached 1.04 mg/m^3 (11:45 A.M.). At midday, the cloudiness decreased, the wind changed direction from south-south-east to south-south-west, and by 12 M., as a result of the development of mixing, the concentration decreased to 0.40-0.30 mg/m^3 at both points. At about 8 P.M., a further decrease of the sulfur dioxide concentration took place when rain fell and the wind turned in a west-southwest direction.

Conclusions

Measurements of sulfur dioxide concentration on the outskirts of Dresden showed that insignificant differences in this concentration exist between a valley and an elevation. The daily variation of sulfur dioxide concentration and daily variation of the frequency distribution in the valley and on the elevation were similar. Differences exist only when the exchange conditions in the valley and on the elevation are different, for example, in stagnant situations and during the passage of fronts. The area of a large city constitutes a voluminous source in the boundary zone of which the sulfur dioxide concentrations show insignificant differences in the horizontal and vertical directions. This was confirmed by measurements at two points separated by a distance of 1.4 km, with a height difference of 120 m.

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H. MROSE, W. WARMBT

REGISTRATION OF SULPHUR DIOXIDE CONTENT IN THE VICINITY OF A CITY — COMPARISON OF MEASUREMENT RESULTS IN A VALLEY AND ON AN ELEVATED PLAIN*

The results of sulphur dioxide concentration measurements in the suburbs of Dresden are stated which were carried out in two points situated in the valley of the Elbe in Radebeul (120m over the sea level) and on an elevated plain near Meteorological Observatory in Wahnsdorf (246m over the sea level) from January to April 1968. The distance between the points amounts to 1.4 km. Continuous registration of sulphur dioxide concentration was done with the help of Novak gas analyzers mounted in these points on the height of 2m. The control of reproducibility of measurement results after the Novak method which was carried out in Wahnsdorf with simultaneous utilization of two analyzers showed a good agreement of data. Divergence of measurement results amounts to 7% on the average, which is probably due to different velocities of solution flow in the instruments.

The results of sulphur dioxide concentration measurement with the help of analyzers were compared with concentrations received by standard method of West-Gaeke. It was found that both methods gave practically similar results. Correlation coefficient between concentrations received by parallel measurements using different methods amounts to about 0.96.

The results of continuous measurements of sulphur dioxide concentration are analyzed in detail at two points. Similar diurnal concentration course was observed, mainly, with maximum about 9 o'clock and minimum about 2 o'clock at both points. The average daily amplitude of sulphur dioxide content amounts to 0.09 mg/m^3 in Radebeul and to 0.07 mg/m^3 in Wahnsdorf. A slight difference in concentrations which amounts, on the average, to about 20% may be explained by possible dilution of sulphur dioxide at a station situated higher up.

Synchronism in the course of concentrations measured at both points is disturbed during the passage of the fronts and wind direction change and also in the presence of stagnant situations in the region of high pressure and formation of local circulation systems. Typical examples of sulphur dioxide concentration registration are cited for various meteorological conditions.

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* Editor's note: The abstract is presented as given in English with the original Russian article.

THEORETICAL AND EXPERIMENTAL STUDY OF THE ASPIRATION COEFFICIENT OF AEROSOLS

S. P. Belyayev, V. M. Voloshchuk, and L. M. Levin (USSR)

From Glavnoe Upravlenie Gidrometeorologicheskoy Sluzhby Pri Sovete Ministrov SSSR. (Chief Administration of the Hydrometeorological Service Under the Council of Ministers of the USSR.) "Meteorologisheskie Aspekty Zagryazneniya Atmosfery". (Meteorological Aspects of Air Pollution.) Sbornik dokladov na mezhdunarodnom simpoziume v Leningrade - Iyul' 1968 g. (Reports delivered at the International Symposium in Leningrad - July 1968.) Pod redaktsiey d-ra fiz.-mat. nauk M. E. Berlyanda. (Edited by Prof. M. E. Berlyand.) Gidrometeorolgicheskoe izdatel'stvo, Leningrad, p. 281-294 (1971). (Hydrometeorological Publishing House, Leningrad, (1971).)

In studying aerodisperse systems, particularly aerosol pollutants of the atmosphere, it is very important to understand the physical processes involved in the aspiration of an aerosol into a given collecting device used for sampling. If the collection is improperly organized, substantial distortions of the concentration and size distribution function of the aerosol particles (particle spectrum) may arise, making the collected sample unrepresentative. The present paper is devoted to the study of these distortions and to the determination of the feasibility of introducing appropriate corrections.

The distortions of aerosol characteristics introduced by the aspiration process may be characterized by the aspiration coefficient ε. For an isodisperse aerosol fraction, it is defined as the ratio of the particle concentration in the collector G to the particle concentration in undisturbed space (flow) G_∞ far from the aspirator.* The difference of ε from unity will determine the degree of destortions caused by the aspiration. In our view, these distortions are chiefly determined by 1) the inertia of the aerosol particles, which causes a difference between the trajectories of aerosol particles and those of particles of the gas medium (ε_i); 2) the deposition of aerosol particles in the connecting lines of the collector (ε_d); 3) the rebound of aerosol particles from parts of the aspirator located close to the intake hole, followed by penetration of these particles into the collector (ε_r). For liquid-particle aerosols, this phenomenon may be substantially complicated by atomization of the droplets. On the whole, the coefficient ε may obviously be represented in the form of the product.

$$\varepsilon = \varepsilon_{\text{и}} \varepsilon_{\text{ос}} \varepsilon_{\text{от}}. \qquad (1)$$

The present paper is devoted to the study of the value of ε_i.

A more or less systematic experimental study of the distortion of an aerosol sample during aspiration was made long ago (Fahrenbach [7]; Zimmerman [8].)

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*For a polydisperse aerosol, one can determine the corresponding mean integral value of the aspiration coefficient.

This question has been treated in many papers, including those of May and Druett [9]; Watson [10]; Walter [11]; Badzioch [12]. Theoretical studies of aspiration were made by Davies [13]; Levin [5], Voloshchuk and Levin [3, 4], and Yur'yev [6].

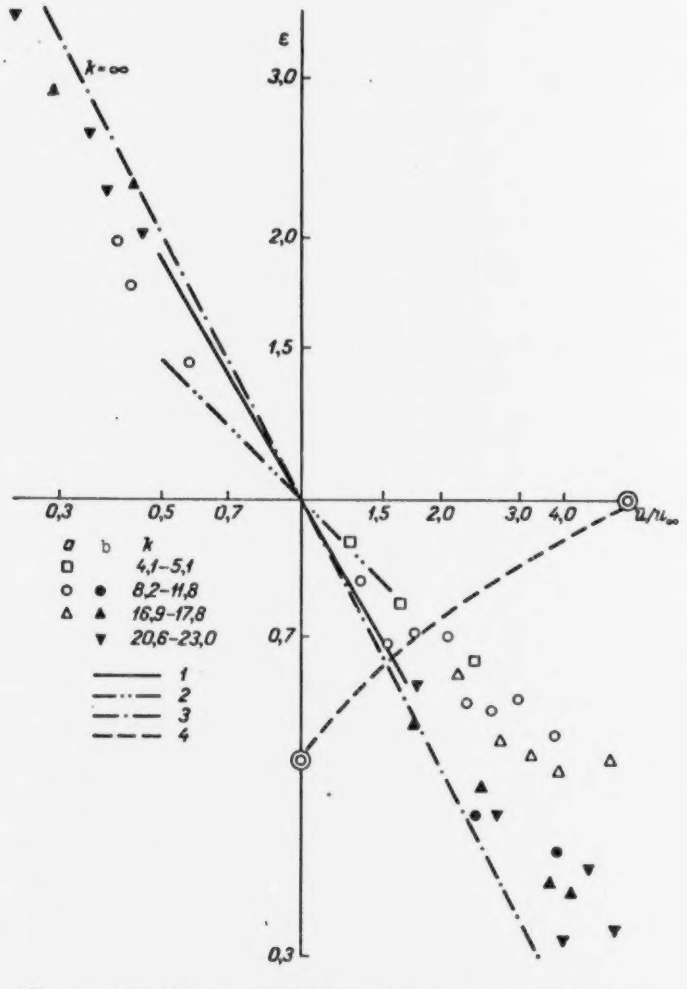

Fig. 1. Comparison of experimental data on the aspiration coefficient k, obtained by different authors (1-4) for zinc (a) and silicate catalyst (b).

It should be noted that experimental studies of aspiration are very contradictory, and frequently cannot be compared since the work was carried out on a polydisperse aerosol, and only the weight concentrations of dust were determined in the sample and in the flow. However, even the results of experiments in which isodisperse aerosols were used are in poor agreement with one another and with theory. As an illustration, Fig. 1 shows values of ε for aspiration into a tube, taken from the papers of Fahrenbach 1), Zimmerman 2), Badzioch 3), and Walter 4). Given here are the dependences of ε on the ratio $\bar{u}/u_\infty$ ($\bar{u}$ being the average velocity in the intake tube and u_∞ the velocity of the incoming flow). Results of experiments with isodisperse aerosols are illustrated with values of the Stokes number k representing the ratio of the inertial forces acting on the aerosol particle to the viscous forces ($k=\frac{u_\infty u_s}{gl}$, where u_s is the particle settling rate, l is the characteristic dimension, for which the tube radius is taken in Fig. 1, and g is the acceleration due to gravity).

As is evident from Fig. 1, when $\bar{u}/u_\infty=1$ (the so-called isokinetic sampling), for a number of experiments $\varepsilon \neq 1$, but Walter gives $\varepsilon \cong 0.5$. In Walter's experiments, even the tendency of ε to change with $\bar{u}/u_\infty$ differs from the results of other authors ($\varepsilon = 1$ when $\bar{u}/u_\infty=5{,}5$).

All this makes the study of aerosol aspiration a timely pursuit aimed at determining ε values, which, by introducing suitable corrections into the characteristics of a collected sample, will make it possible to obtain a correct representation of the characteristics of the aerosol studied.

1. Theoretical Studies of Aspiration

Theoretical papers on aspiration deal with the determination of the aspiration coefficient ε_i, which may also be defined as the ratio j of a flow of particles, through any surface $\hat{S}$ imagined in the region of the stream, to the flow through this surface of inertialess particles moving far from the aspirator in the same manner as inertial particles:

$$\varepsilon_i = \iint\limits_{\hat{S}} \mathbf{j}(k)\, dS \Big/ \iint\limits_{\hat{S}} \mathbf{j}(0)\, d\hat{S}. \qquad (2)$$

In the general case, the magnitude of ε_i depends on the Stokes number and other parameters

$$\varepsilon_i = \varepsilon_i\left(k;\ \frac{\bar{u}}{u_\infty};\ \hat{S};\ L_i;\ \mathrm{Re};\ \mathrm{Re}^*;\ \ldots\right). \qquad (3)$$

where L_i are some geometric flow parameters (for example, when the aerosol is sucked into a tube, the ratio of its inner diameter d to the outer diameter D, etc.), and Re and Re* are the Reynolds numbers of the particle and flow respectively.

To find ε_i, it is necessary to solve two difficult problems involving the determination of the field of velocities of inertialess particles u(r) and field of velocities and concentrations of aerosol particles moving in the field u(r). Since a detailed discussion of the procedures involved is beyond the scope of this paper, we refer the reader to [5, 3, 4, 6]. We will here confine ourselves only to a description of the results obtained.

Aspiration of aerosol particles into a tube (axisymmetric problem) or slit (two-dimensional problem) from a flow parallel to the walls of the tube or slit was studied in [3, 4]. This involved a discussion of two extreme types of streams of the gaseous phase in the tube (slit), unseparated and separated (Kirchhoff type), characterized by inertial separation of flow from the edges of the tube (slit). Figure 2a shows diagrams of stream lines for unseparated flows, and Fig. 2b, for separated ones (the shaded area represents the so-called dead zone, i. e., rest zone separated from the flow by a free boundary). Obviously, both diagrams constitute mathematical models of flows, and the actual flow must differ from them. However, it is our impression that the actual flow will constitute something intermediate between the above models, since in the first case the influence of the wake is completely neglected, and in the second case, the strongest wake apparently develops as compared with the real wake.

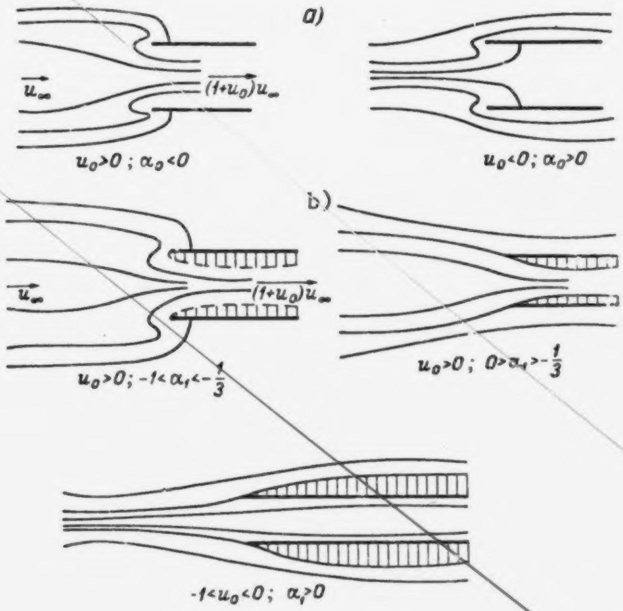

Fig. 2. Stream patterns.

As the dimensionless velocity field for the two-dimensional case (slit) we took the velocities in two-dimensional Bord mouthpieces which on the complex plane z = x + iy are described by the formulas

$$u = 1 + u_0\xi; \quad \pi z = -\frac{1}{\xi} - \ln\left(\frac{1}{\xi} - 1\right); \tag{4}$$

$$u = 1 + u_0\xi; \quad \pi z = \ln\frac{2}{\sqrt{e}} - \frac{1}{2\xi} - \ln\left(\frac{1}{\xi} - 1\right); \tag{5}$$

where the complex velocity $u_x = u_x - iu_y$, and ξ is a complex parameter. Equation (4) describes the unseparated flow in a Bord mouthpiece (Fig. 2a) on which impinges from infinity a flow parallel to the abscissa axis at velocity $u_\infty = 1$ when $x = -\infty$ and velocity u = 1 + u_0 inside the Bord mouthpiece when $x \to +\infty$. Similarly, Eq. (5) describes the corresponding separated flow. In describing the velocity field in an axisymmetric tube (Bord mouthpiece), use was made of the numerical solution to the problem, obtained by Vandrey [14] and describing separated flow.

The ratio $\overline{u}/u_\infty$ is commonly considered for characterizing the anisokineticity of a flow in a tube (slit). For theoretical formulas, it is more convenient to use the anisokineticity coefficient

$$\alpha = -1 + \frac{1}{\overline{u}} = -1 + \frac{l^\beta u_\infty}{Q}. \tag{6}$$

Which depends uniquely on this ratio. Here Q is the discharge of the gaseous medium through the aspirator. To unify the formulas, the quantities referring to unseparated potential flow past a slit are denoted by subscript 0; to separated flow past a slit, by subscript 1, and to separated flow past a tube, by subscript 2.

If $u_0 < 0$, the flow within a tube (slit) is decelerated, and the wake forms outside the aspirator. Then, in all three cases,

$$\overline{u} = 1 + u_0 \quad \text{и} \quad \alpha = -1 + \frac{1}{1 + u_0}. \tag{7a}$$

When $u_0 > 0$ (the flow within the aspirator is accelerated)

$$\overline{u}^{(0)} = 1 + u_0, \quad \alpha = -1 + \frac{1}{1 + u_0}; \tag{7b}$$

$$\overline{u}^{(s)} = 1 + 0{,}5u_0, \quad \alpha_s = -1 + \frac{1}{1 + 0{,}5u_0}, \quad s = 1, 2. \tag{7c}$$

The presence of the multiplier 0.5 before u_0 in (7c) is due to the fact that in separated flow of the type under consideration, the region of motion of the carrying medium in the aspirator occupies only a certain middle portion, so that when

$$x \to \infty, \quad y_\infty^{(s)} = \left(\frac{1+0{,}5u_0}{1+u_0}\right)^{1/s} \quad (s=1,2).$$

The following asymptotic formulas* were used to determine the aspiration coefficient for the entire slit or tube in [3] and [4]:

$$\varepsilon_s(k) = \varepsilon_s^{(0)}(k) + \begin{cases} 0\left\{\left(\frac{k}{k_{cr}}\right)^{-2}\ln^2\left(\frac{k}{k_{cr}}\right)\right\} & s=0,1 \\ 0\left\{\left(\frac{k}{k_{cr}}\right)^{-2}\right\} & s=2 \end{cases} \tag{8a}$$

where the asymptotic value (for $k \to \infty$) of $\varepsilon_s(k)$ is

$$\varepsilon_s^{(0)}(k) = 1 + \alpha_3 E_s(k), \quad s = 0, 1, 2, 3 \ldots \tag{8b}$$

$$E_0(k) = \begin{cases} 2e^{-2} & k=(2\pi)^{-1} \\ \left(\frac{k}{\pi}\right)^{-\frac{\pi}{k}}\left[\Gamma\left(1+\frac{\pi}{k}\right)e^{\frac{\pi}{k}}\right]^{-1}, & k \geqslant \frac{1}{\pi} \end{cases} \tag{8c}$$

$$E_1(k) = \begin{cases} 4\left(3e^{-2} - e^{-1}\right), \; k=(2\pi)^{-1} \\ 2^{\frac{\pi}{k}}e^{-\frac{\pi}{2k}}\left\{-1 + 2k\sin\frac{1}{k}\Gamma\left(1-\frac{\pi}{k}\right)\times\right. \\ \left.\times\left[e^{-\frac{\pi}{2k}}2^{-\frac{\pi}{k}}\frac{k}{\pi}^{-\frac{\pi}{k}} + \frac{\pi}{2k}\Gamma\left(\frac{\pi}{k};\; \frac{\pi}{2k}\right)\right]\right\}, \; k \geqslant \frac{1}{\pi} \end{cases} \tag{8d}$$

$$E_2(k) = 2{,}825 - \frac{4{,}575}{1+k} + \frac{1{,}750}{1+2k} - \left(1{,}936 - \frac{3{,}718}{1+k} + \right.$$
$$\left. + \frac{1{,}768}{1+2k}\right)e^{-\frac{0{,}2}{k}} + \left(0{,}111 + \frac{0{,}044}{1+k} + \frac{1{,}627}{1+2k}\right)e^{-\frac{2}{k}}. \tag{8e}$$

Table 1 gives values of $E_s(k)$, and the corresponding values of E_s for different u/u_∞ (or α), and k are given in Table 2.* The asymptotic values of local aspiration coefficients near the axis of symmetry of the slit or tube were found in the same manner. Analysis of the results obtained shows that: a) the aspiration coefficient for the tube (slit) is by-and-large linearly related to the anisokineticity coefficient α (therein lies the advantage of introducing α); b) the effect of the wake on the aspiration

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*By k_{cr} is meant a value of the Stokes number for which, near the edges of the tube or slit there takes place a total "quenching" of the uniform linear motion that the particles execute in an undisturbed flow (for more detail, see [5, 2]).

Table 1

Values of the Functions $E_s(k)$

πk	$E_0(k)$	$E_1(k)$	$E_2(k)$	πk	$E_0(k)$	$E_1(k)$	$E_2(k)$
0,5	0,270	0,153	0,19	5,0	0,646	0,590	0,79
1,0	0,370	0,258	0,32	10,0	0,755	0,720	0,91
2,0	0,484	0,395	0,51		1,000	1,000	1,00

Table 2

Values of $\varepsilon_s(k)$ for Different s, α and k
(for a given α, the first rows correspond to s = 0, the second rows to s = 1, and the third rows, to s = 2)

α	πk						u_0	$\pi k_{\text{кр}}$
	0,5	1,0	2,0	5,0	10,0	∞		
−0,80		0,704	0,613	0,483	0,396	0,2	4,0	0,800
		0,793	0,684	0,528	0,424	0,2	8,0	0,778
		0,744	0,592	0,368	0,272	0,2	8,0	
−0,60	0,838	0,778	0,710	0,612	0,457	0,4	1,5	0,225
	0,908	0,845	0,763	0,646	0,568	0,4	3,0	0,187
	0,886	0,808	0,694	0,526	0,454	0,4	3,0	
−0,40	0,892	0,852	0,806	0,741	0,698	0,6	0,667	0,067
	0,939	0,897	0,842	0,764	0,712	0,6	1,333	0,024
	0,924	0,872	0,796	0,684	0,636	0,6	1,333	
−0,20	0,946	0,926	0,903	0,871	0,949	0,8	0,250	0,012
	0,969	0,948	0,921	0,882	0,856	0,8	0,500	
	0,962	0,936	0,898	0,842	0,818	0,8	0,500	
+0,25	1,068	1,093	1,121	1,162	1,189	1,25	−0,200	0,012
	1,038	1,064	1,099	1,148	1,180	1,25	−0,200	
	1,048	1,080	1,128	1,198	1,228	1,25	−0,200	
+0,50	1,135	1,185	1,242	1,323	1,378	1,50	−0,333	0,042
	1,076	1,129	1,198	1,295	1,360	1,50	−0,333	
	1,095	1,160	1,255	1,395	1,455	1,50	−0,333	
+1,0	1,270	1,370	1,484	1,646	1,755	2,0	−0,500	
	1,153	1,258	1,395	1,590	1,720	2,0	−0,500	
	1,190	1,320	1,510	1,790	1,910	2,0	−0,500	
+2,0	1,540	1,740	1,968	2,292	2,510	3,0	−0,667	0,332
	1,306	1,516	1,790	2,180	2,440	3,0	−0,667	
	1,380	1,640	2,020	2,580	2,820	3,0	−0,667	
+4,0		2,480	2,936	3,584	4,020	5,0	−0,800	0,800
		2,032	2,580	3,360	3,880	5,0	−0,800	
		2,280	3,040	4,160	4,640	5,0	−0,800	

coefficient decreases with increasing Stokes number k and increases in direct proportion to the absolute value of the anisokineticity coefficient α; c) for $k \sim (10\text{-}100)k_{cr}$, the aspiration coefficient differs markedly from both $\varepsilon(k=\infty)=1+\alpha$ and $\varepsilon(k=0)=1$. For example, when $\alpha \sim -0{,}5$ with $k \approx 10k_{cr}$, $\frac{1}{2}[\varepsilon_0^{(0)}+\varepsilon_1^{(0)}) \sim 0{,}84$, and when $k \sim 100k_{cr}$, $\frac{1}{2}[\varepsilon_0^{(0)}+\varepsilon_1^{(0)}] \sim 0{,}63$, whereas in this case $\varepsilon(\infty)=0{,}5$; d) in many cases, the influence of the wake on ε may be neglected. For example, when $|\alpha| \leqslant 0{,}5$ and $k > 0.3$, the difference $\varepsilon_0^{(0)} - \varepsilon_1^{(0)}$, referred to $\frac{1}{2}[\varepsilon_0^{(0)}+\varepsilon_1^{(0)}]$, amounts to less than 4%; e) other things being equal, when $\alpha<0$, $\varepsilon(k)$ for a tube is less than for a slit, and when $\alpha>0$, vice versa, i. e., for the axisymmetric case, ε is closer to unity than to the corresponding planar case; f) the aspiration coefficient for the central portion of aspirators may differ substantially from the total coefficient. For s = 0, this difference is much greater than for s equal to 1 and 2. The formation of a wake with decreasing inhomogeneity of the velocity field in the entrance portion of the aspirator decreases the inhomogeneity of the local aspiration coefficient.

For large values of u_0, formulas (8) do not solve the problem. For this case, the small parameter method was used to find values of the aspiration coefficient for a slit in the form of a power series in $k' = \frac{\pi k}{\bar{u}}$:

$$\varepsilon_s(k') = 1 - \mu_s k' + 0\left(k'^2\right), \quad s = 0, 1, \quad k' \to 0, \tag{9a}$$

$$\mu_0 = \frac{1}{\pi}\alpha^2 \int_0^\pi \alpha\nu \frac{\sin^2\alpha}{\nu^2} \frac{1-(1+\alpha)\frac{\sin^2\nu}{\nu^2}}{\left[1-(1+\alpha)\frac{\sin 2\nu}{\nu}+(1+\alpha)^2\frac{\sin^2\nu}{\nu^2}\right]^2},$$

$$\mu_1 = \frac{4}{\pi}\alpha^2\frac{1}{(1+\alpha)^2}\int_0^\pi d\nu\left\{-\frac{2\pi}{1+\alpha}+\theta-\frac{1-\alpha}{1+\alpha}\nu\right\}^{-2}\sin^2\theta\,\frac{d\theta}{d\nu}, \tag{9b}$$

where $\theta(\nu)$ satisfies the transcendental equation

$$\theta(\nu) = \pi + \operatorname{arc\,tan}\left\{-\operatorname{ctn}\frac{\nu-\theta}{2}-\left[\frac{2\pi}{1+\alpha}+\theta-\frac{1-\alpha}{1+\alpha}\nu\right]^{-1}\right\}. \tag{9c}$$

For s = 0, formulas (9) hold for the entire interval of variation of α, i. e., for $-1 \leqslant \alpha < \infty$, and when s = 1, only for $-1 \leqslant \alpha \leqslant \leqslant -\frac{1}{3}$. The values of μ_0 and μ_1 for some values of α are given in Table 3. For $\alpha \to -1+0$, the following asymptotic relation holds:

$$\mu_1 = \alpha^2\{0{,}451 + 0{,}664(1+\alpha) + 0[(1+\alpha)^2]\}. \qquad (10)$$

Table 3

Values of the Functions $\mu_o(\alpha)$ and $\mu_1(\alpha)$

α	u_0	μ_0	α	u_0	μ_0
−1,00	∞	0,454	+0,05	−0,048	−0,242
−0,95	19	0,440	+0,10	−0,091	−0,233
−0,90	9	0,428	+0,20	−0,167	−0,221
−0,80	4	0,405	+0,40	−0,286	−0,199
−0,70	2,333	0,383	+0,60	−0,375	−0,182
−0,60	1,5	0,362	+0,80	−0,444	−0,168
−0,50	1,0	0,342	+1,00	−0,500	−0,156
−0,40	0,667	0,322	+1,50	−0,600	−0,133
−0,30	0,429	0,303	+2,00	−0,667	−0,117
−0,20	0,250	0,285	+2,50	−0,714	−0,104
−0,10	0,111	0,267	+3,00	−0,750	−0,945
−0,05	0,053	0,258	+3,50	−0,778	−0,0865
0	0	0	+4,00	−0,800	−0,0800

α	−0,85	−0,80	−0,75	−0,70	−0,65	−0,60	−0,55	−0,50	−0,45
u_0	11,333	8,000	6,000	4,667	3,714	3,000	2,444	2,000	1,636
μ_1	0,413	0,402	0,389	0,374	0,359	0,344	0,325	0,306	0,284

Comparison of (9) and (10) with the formula

$$\varepsilon(k') = 1 - 0{,}451k' - 0{,}148k'^2 + 0(k'^3), \qquad (11)$$

obtained in [8] shows that when $\alpha \to -1+0$, they coincide asymptotically. If $-1 \leqslant \alpha \leqslant -0{,}7$, i. e., $\overline{u}/u_\infty > 3{,}3$, formulas (9) and (11) coincide to within 15%. For the axisymmetric case, the following value was obtained in [5] :

$$\varepsilon(k'') = 1 - 0{,}80k'', \qquad (12)$$

where

$$k'' = \frac{2k}{\sqrt{\overline{u}}}.$$

2. Experimental Studies of Aspiration

The experimental study of the aspiration process was carried out on a special stand that made it possible to photograph the tracks of aerosol particles aspirated into a cylindrical tube from a flow. The stand consisted of a closed-type wind tunnel in which the intake tube studied was mounted. By blowing in isodisperse particles (spores of various plants were chiefly used) at a certain distance from the tube (ahead of the flow), one can obtain an aerosol flow that is partially sucked into the tube. Two flash lamps located in a vertical plane illuminated in the flow a parallelepiped measuring 53 x 80 x 1.5 mm. The flow was photographed in the direction of the 1.5 mm side. Patterns of flow of aerosol particles past the vertical meridional plane of the intake tube were thus obtained. The photographs were taken under two sets of illumination conditions: 1) when the pulses were relatively long (of the order of 10 msec), and 2) when the illumination consisted of three short consecutive pulses (of the order of 25-200 μsec).

In the first case, it was convenient to determine the particle trajectories from a photograph, and in the second, from the distance between the particle track lines one could determine the velocity field of the particles. A detailed description of this stand is given in [1]. This stand can be used to study both aspiration and deposition of aerosol particles from a flow. The study was made on tubes up to 2.5 cm in dismeter at flow velocities from 2 to 20 m/sec with particles from 10 μm in diameter.

By determining on the photographs the limiting paths of aerosol particles, which are defined by the particles entering the tube and flowing past it, one can measure the tube diameter of these limiting trajectories Φ in the unperturbed region of the flow (at a distance of 5-6 tube diameters), and use it to determine the aspiration coefficient

$$\varepsilon_i = \frac{G}{G_\infty} = \frac{\Phi^2 u_\infty}{d^2 \bar{u}}. \qquad (13)$$

The value of $\bar{u}$ was determined from measurements of the flow rate of air through the aspiration tube. The errors thus determined by the aspiration coefficient ε_i did not exceed 10%.

In order to be able to compare the experimental and theoretical data, we first conducted experiments with a very thin-walled tube (the wall thickness was 1% of the inner tube diameter α).

Results obtained from the study of the dependence of ε on $\bar{u}/u_\infty$ for Stokes numbers of 0.8, 1.2 and 5.2 and for the thin-walled tube are shown in the graph of Fig. 3. For comparison, the same graph gives the theoretical curves for the

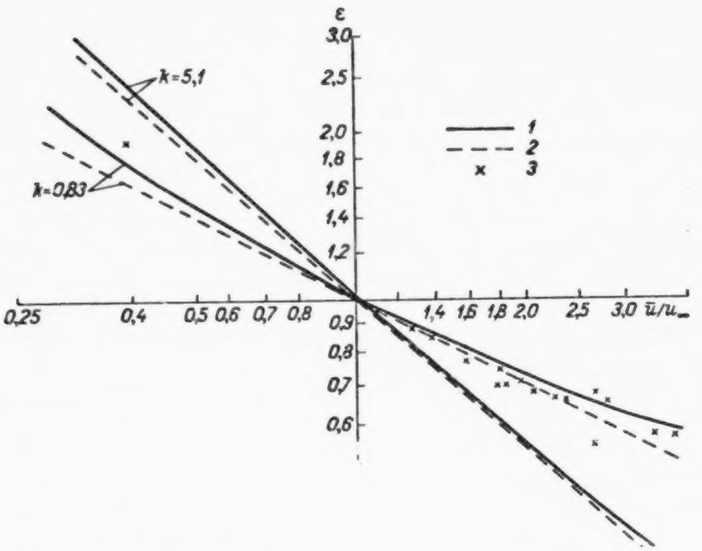

Fig. 3. Theoretical (1) and experimental (2) curves of aspiration coefficient ε_i for thin-walled tubes, and Badzioch's data for $x = 5.1$ (3).

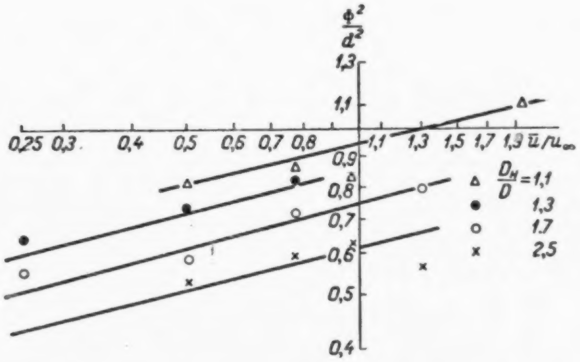

Fig. 4. Measured values of $\frac{\Phi^2}{d^2}$ for thick-walled tubes (Stokes number $k = 1.24$).

corresponding Stokes numbers. It should be noted that the agreement of the theoretical and experimental data is fully satisfactory, and as the Stokes number increases, the difference between these two kinds of data decreases.

In order to determine the influence of the wall thickness on the aspiration, a special series of experiments were carried out with tubes having a relative wall thickness h/d equal to 0.02, 0.3, 0.7, and 1.5 (here h is the absolute tube-wall thickness). The results obtained are illustrated in Fig. 4, with data of one of the series of experiments. As shown in Fig. 4, for a given number k, the graphs of the functions Φ^2/d^2 versus $\overline{u}/u_\infty$ in log-log coordinates are straight lines whose slope is independent of the tube wall thickness (to a first approximation). In addition, even for an isokinetic collection of the sample, $\varepsilon_i \neq 1$. Thus, if the wall thickness is substantially different from 0, the sample collected under isokinetic conditions will not be representative. Strictly speaking, for a tube with thick walls and for a polydisperse system, the sample will not be representative at any intake velocity, since for the given values of $\overline{u}/u_\infty|$ one can have $\varepsilon = 1$ for only one value of k.

Thus, in the range $0{,}2 < k < 20$ when the sample is collected from the aerosol flow, to minimize the difference between the characteristics of the sample and the aspirated aerosol, two conditions are required: it is necessary to provide for isokinetic conditions of collection and to construct a tube with the thinnest possible wall thickness. Otherwise, suitable corrections must be introduced.

It may be postulated that the disagreement of the experimental results (see Fig. 1) is due to the fact that in different experiments, the wall thickness of the actual tubes was different and determined different ε_i values. However, a study of the photographs of the process of aspiration into thick-walled tubes showed that substantial rebounding of the particles off the end plane of the tube walls is possible, and that most of the particles are sucked into the tube after rebounding. Obviously, in a gravimetric determination of the concentration (used by all the authors of the papers cited), all the particles that have entered the tube as a result of rebounding off its end plane become suspended in the total sample with the remaining particles.

In our experiments, however, we studied the "purely inertial" aspiration (determined by the interaction of inertial and hydrodynamic forces), and particles that rebounded off the end plane of the tube were not considered. Apparently, this as well as the fact that the other authors consider the influence of certain other secondary effects (for example, the deposition of particles inside the tube) explains the discrepancy in the experimental results. As proof of this statement, we will cite the results obtained in one of the most comprehensive studies of Badzioch [12] (see Fig. 1). However, in contrast to the work of Badzioch, we give a different notation to the points obtained for the same Stokes number but differing only in the mechanical properties of the particles. It is obvious that for the same values of $\bar{u}/u_\infty$ and k, the aspiration coefficient ε_1 should assume a completely defined value regardless of the particle material. As is evident from Fig. 1, in Badzioch's experiments, the results obtained for silicate catalyst particles differ markedly from those obtained for zinc particles. Apparently, this unambiguously confirms the presence of a substantial rebounding of the particles off the leading edge of the tube. It may be postulated that particles that have rebounded off the silicate catalyst are sucked in to a greater extent than zinc particles. Moreover, using data obtained by photographing the aspiration process, one can state that the so-called dead zones obtained by Walter [11] by photographing aspiration with a long exposure are but the image of particles of incoming aerosol that rebounded off the tube during the exposure. For this reason, the conclusions reached by Walter should not be applied to pure inertial aspiration, but to aspiration complicated by other secondary effects. These conclusions are valid only for a given tube (of a given shape), given particles (of given material) and given aspiration conditions.

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S. P. BELYAEV, V. M. VOLOSHTCHUK, L. M. LEVIN

THEORETICAL AND EXPERIMENTAL INVESTIGATIONS OF ASPIRATION COEFFICIENT*

The methods of evaluation and experimental measurement of aspiration coefficients are worked out for slots and tubes which determine the relation of aerosol particle concentration in inflowing current and in aspiration mechanism.

Aspiration coefficients for large and small numbers k are calculated under potential flow patterns with and without breakaway for a case of Bord flat nozzle. Interpolation for mean Stocks numbers is carried out and applicability boundaries of obtained results are shown. The influence of wake and anisokinetic motion of aerosol particles on aspiration coefficient is analysed. Analogous results are received for Bord axis-symmetric nozzle (a model of axis-symmetric thin-walled tube) under potential flow pattern with breakaway. Aerosol aspiration into a slot situated inside a flat canal is investigated.

Aspiration coefficients for tubes with different relative thickness of the walls h/D are received with the help of track photographing method of isodispersed aerosol particles. It was found that aspiration coefficient E and ratio u/u_∞ are connected mutually by inverse power law

$$E = A(h/D,\ k)(\bar{u}/u_\infty)^{-c(k)},$$

where u is mean velocity in the tube, u_∞ is velocity of inflowing current, while $A(u/D,\ k) \to 1$ at $h/D \to 0$ and is decreased with increase of h/D. The comparison of experimental and theoretical data carried out for little h/D (thin-walled tubes) showed good convergence. The obtained results, both theoretical and experimental differ substantially from experiment data of Badzioch and Walter and are in bad agreement with Watson data. This can be explained by the fact that we investigate aspiration determined only by interaction of inertial and hydrodynamic forces which is not complicated by the occurrence of particle break off from the edges and their sedimentation on tube walls.

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*Editor's note: The abstract is presented as given in English with the original Russian article, but has been slightly edited.

MECHANISM OF PHOTOCHEMICAL POLLUTION OF THE URBAN ATMOSPHERE

M. T. Dmitriyev, N. A. Kitrosskiy, and V. A. Popov (USSR)

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Photochemical reactions in atmospheric air are of interest from the geophysical as well as the sanitary-hygienic point of view. Of the 26 gases occurring in pure air, in addition to oxygen, photochemical reactions may involve the participation of nitrogen oxides, ozone, sulfur dioxide, formaldehyde, ammonia, and halogens. However, these substances are present in very slight amounts whose sum total does not exceed 0.3 mg/m^3, so that the photochemical reactions involving them are not in any way significant.

Of practical importance may be only the photochemical formation of ozone in the surface layer. Nevertheless, analysis of the daily variation of ozone concentrations associated with meteorological conditions shows that of the four main sources of ozone formation, the most effective process is the photochemical oxidation of oxygen on clear days in autumn or spring, due to the penetration of short-wavelength radiation into the troposphere (with the ozone concentrations reaching 50-80 μg/m^3 in some cases). The most frequent increases in ozone concentrations (to 100-150 μg/m^3) are due to penetration of air masses from the lower stratosphere. Among other sources of ozone may be mentioned its formation preceding storms, 2 to 4 hours before precipitation [3], during which the concentrations reach 200-250 μg/m^3, and the formation of ozone during lightning flashes, snowfalls, and snowstorms.

Under natural conditions, the photochemical formation of ozone takes place most frequently during oxidation of tree resins in coniferous forests, or during oxidation of seaweed thrown up onto the shore, this being associated with an increase in ozone concentrations to 50-60 μg/m^3. However, it is obvious that all natural processes of ozone formation in the surface layer, which undoubtedly are of practical hygienic importance, may be completely neglected in comparison with the photochemical formation of ozone in the polluted atmosphere of cities in which the ozone concentrations reach 2 mg/m^3 or more [6].

Industrial emissions and motor transport considerably increase the number of secondary components of atmospheric air, and this in turn increases the quantity of photochemical reactions taking place in air and sharply increases their effectiveness. Studies of the composition of atmospheric pollutants show that as a rule, air contains many more chemical substances than are present in the emissions of chemical plants or motor transport. This is due to numerous chemical reactions occurring in atmospheric air. Under the influence of physicochemical factors, chiefly short-wavelength radiation, atmospheric pollutants undergo various transformations through photolysis (upon irradiation with sunlight), ozonolysis (in reactions with ozone), hydrolysis (in reactions with water vapor), pyrolysis (decomposition on heating), oxidation (reaction with oxygen), etc. Moreover, atmospheric pollutants and the new chemical substances formed enter into reactions between themselves, so that the quantity of products of these reactions is greatly increased.

In a sanitary-hygienic sense, the reactions of greatest interest are physicochemical ones, which form new atmospheric pollutants whose toxicity substantially exceeds that of the initial compounds. Thus, exhaust gases form acrolein, aldehydes, ketones, organic peroxides. As the molecular weight of the compound formed increases, its concentration decreases, but the toxicity rises sharply. A particularly toxic compound formed in the atmosphere and first observed in urban air is peroxyacetyl nitrate (molecular weight, 121.05) and other compounds of its homologous series [6]. This type of processes also includes the formation of the so-called photochemical smog in the air of large cities and industrial centers. In this case, in addition to a strong biological effect (acute irritation of the upper respiratory tract and eyes, aggravation of asthmatic diseases, etc.), there is a sharp decrease in visibility. For this reason, in the presence of a high pollution level and a sufficiently intense solar radiation (above 0.5 cal/cm^2 min), large American cities appear to be engulfed in a yellowish blue fog [6].

However, both laboratory and full-scale studies show that the formation of photochemical smog takes place gradually, in the absence of obvious indications (mist, acute irritation of the eyes), with urban air always containing the same compounds as are present in smog. The main components of photochemical smog are photooxidants (ozone, organic peroxides, nitrate and nitrite, compounds of the peroxyacetyl nitrate series), nitrogen oxides, carbon monoxide and dioxide, hydrocarbons (alkanes, alkenes, aromatics), aldehydes, ketones, phenols, methanol, ketene, epoxides, various radicals, etc. In an intense photochemical smog, in the air of American cities, the concentrations of ozone reach 2 mg/m^3 (20 times the MPC of work areas), those of nitrogen oxides, 6 mg/m^3 (60 times the mean daily MPC for atmospheric air), while the hydrocarbon concentrations reach 1.5-2 g/m^3 [6].

The influence of meteorological conditions on chemical reactions in atmospheric air substantially complicates their investigation. In this connection, simulation methods assume a major importance in the study of this problem under laboratory conditions. The purpose of simulation is to study the chemical mechanism of the processes and to create concentrations of oxidants and other newly formed pollutants that correspond to photochemical smog. In particular, we used the photochemical, electronic-chemical, electric-discharge and thermochemical methods. For example, ozone and nitrogen oxide concentrations characteristic of photochemical smog are formed by passing pure air across an electric field. The simulation method was also used to study the formation of heavy ions during the combustion of various substances, the action of electrons on air containing nitrogen oxides, ozone, olefins, vapors of ethanol, acetone, acetaldehyde, tetrahydrofuran, hexane, dioxane, heptane, nitrobenzene, etc.

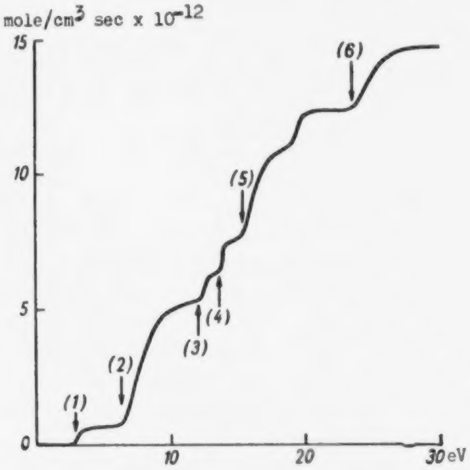

Fig. 1. Dependence of overall reaction rate of formation of ozone and nitrogen oxides in pure air (mol/cm^3 sec x 10^{-12}) on (as a function of) electron energy.

Photochemical reactions in air were studied, particularly in the presence of substances that are the most frequent atmospheric pollutants. Because of the experimental difficulties involved in the determination of the threshold energies of photochemical processes, the irradiation of air in the various experiments was conducted at a pressure of the order of 0.1 Torr with slow electrons whose energy was varied by an electric field (method of critical potentials).

Fig. 1 shows the overall reaction rate as a function of the electron energy, obtained for pure air. The curve was obtained by differentiating the experimental dependence of pressure in a closed vessel on the electron energy, the reaction products being diverted onto a cooled surface, and for this reason each consecutive rise of the curve corresponds to the critical potential of a new reaction.

From the data obtained one can draw a conclusion concerning the course of the following photochemical reactions (the numbers of which correspond to the numbers given in Fig. 1) in pure air and with the corresponding threshold energies and wavelengths of light:

Process	Electron Energy, eV	Wave-length, nm
(1) $O^- + O_2 + M \rightarrow O_3^- + M$	3,0	407
(2) $O_2^*(^3\Sigma_u^-) + O_2 \rightarrow O_3 + O$	6,1	200
(3) $O_2^+ + N_2 \rightarrow NO^+ + NO$	12,2	100.
(4) $O^+ + N_2 \rightarrow NO^+ + N$	13,5	90
(5) $N_2^+ + O_2 \rightarrow NO^+ + NO$	15,6	78
(6) $N^+ + O_2 \rightarrow NO^+ + O$	24,0	51

Below 3 eV (407 nm) no reaction takes place in pure air, and as the energy increases, the formation of ionized ozone is observed, caused by the appearance of negative ions of O-atomic oxygen (reaction 1). However, this process takes place only in the presence of an excess of free electrons, and therefore it is practically undetectable in the presence of ultraviolet radiation. An intense formation of ozone takes place at an electron energy of 6.1 eV (200 nm), which corresponds to the appearance of excited oxygen molecules that are not yet able to dissociate (reaction 2). When positive oxygen ions appear at 12.3 eV (100 nm), a slight formation of nitric oxide begins that is then somewhat intensified during ionization of atomic oxygen at 13.5 eV or 90 nm (reaction 4). An intense formation of nitric oxide takes place during ionization of molecular nitrogen at 15.7 eV or 78 nm (reaction 5). The formation of nitric oxide is substantially accelerated during the dissociative ionization of nitrogen at 24 eV or 51 nm (reaction 6). The method of critical potentials was also used to study the mechanism of chemical reactions in air in the presence of nitrogen oxides and ozone.

Fig. 2 shows the rate of formation of ozone and nitrogen oxides as a function of the electron energy in air in the presence of nitrogen oxides and ozone. Whereas ozone is formed in air in accordance with reaction (2) at an electron energy of 6.1 eV (curve 2), the formation of ozone in a mixture of air and nitric oxide begins only at an electron energy of 2.1 eV or 580 nm (curve 3). A much more intense formation of ozone takes place in the reaction of nitrogen dioxide with oxygen at an electron energy of 3.1 eV (curve 1), which corresponds to a wavelength of light of 395 nm. At an electron energy of 7 eV, the reaction rate of ozone formation in the presence of nitrogen dioxide is 5-6 times as high as in pure air. The formation of nitric oxide, which begins in accordance with reaction (5) at 15.6 eV (curve 4), takes place to the same extent in pure air and in the presence of ozone (black points). The formation of ozone in the presence of nitric oxide and nitrogen dioxide may be explained by the excitation, dissociation or reaction of nitrogen oxide molecules and the secondary reaction of atomic oxygen:

$$NO^{*} + O_2 \rightarrow NO_2 + O, \tag{7}$$

$$NO_2 - \wedge\!\!\wedge \rightarrow NO_2^{\bullet} \rightarrow NO + O, \tag{8}$$

$$NO_2^{\bullet} + O_2 \rightarrow NO + O_3, \tag{9}$$

$$O + O_2 + M \rightarrow O_3 + M. \tag{10}$$

It is interesting to note that the formation of ozone from nitric oxide begins at a lower electron energy than from nitrogen dioxide. However, in the latter case the reaction rate is higher. This fact may be accounted for to some extent by the influence of pressure on reaction (2). Whereas the formation of ozone from the dioxide is a molecular reaction (reaction 9), the rate of ozone formation from nitric oxide is almost completely determined by a slower termolecular reaction (reaction 10). Thus it may be expected that at atmospheric pressure, at which the termolecular process is more probable, the rate of photochemical reactions of ozone formation from nitric oxide is not very different from the reaction in the presence of nitrogen dioxide. Under the influence of radiation of ultraviolet lamps, the formation of ozone takes place at wavelengths up to 253 nm (1), but the main reaction with a quantum yield of about 2, corresponding to reactions (2 and 10), takes place at 180-220 nm, which is in accord with reaction (2), recorded during the action of electrons. At wavelengths above 200 nm, the quantum yield quickly decreases to 0.1 molecule per absorbed quantum. For the 180-253 nm wavelength range of the spectrum of PRK (straight mercury-quartz) gas-discharge lamps, the energy yield of ozone in air amounts to an average of 22 molecules per 100 eV, or 395 g/kW-hr (for a quantum yield of 1.2).

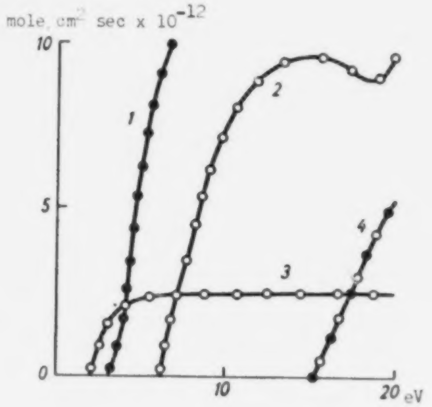

Fig. 2. Reaction rate in the presence of nitrogen oxides and ozone versus electron energy.

1 - formation of ozone in a mixture of air and nitrogen dioxide, 2 - in pure air, 3 - in a mixture of air and nitric oxide, 4 - formation of nitric oxide in pure air and in the presence of ozone (black points).

Fig. 3 gives values of the energy yield of ozone formation during the action of the PRK lamp, mounted at the center of the chamber, as a function of its volume. Inasmuch as the degree of absorption is small (less than 1%) when the dimensions of the chamber are of the order of a few meters, and the yield of ultraviolet radiation does not exceed 15%, the ozone yields obtained, amounting to 2-4 g/kW-hr of the lamp's energy, are considerably below the photochemical energy yield.

Additional studies have established that the formation of nitric oxide and nitrogen dioxide under the influence of radiation in the 180-253 nm wavelength range initially occurs via ozone, and later, mainly via nitrous oxide, which is a product of a termolecular reaction that is not detected because of its low rate at low pressures.

The following reactions take place in this case:

$$O_3 \rightsquigarrow O_2 + O, \tag{11}$$

$$N_2 + O + M \rightarrow N_2O + M, \tag{12}$$

$$N_2O + O \rightarrow 2NO, \tag{13}$$

$$NO + O_3 \rightarrow NO_2 + O_2. \tag{14}$$

$$NO + O + M \rightarrow NO_2 + M. \tag{15}$$

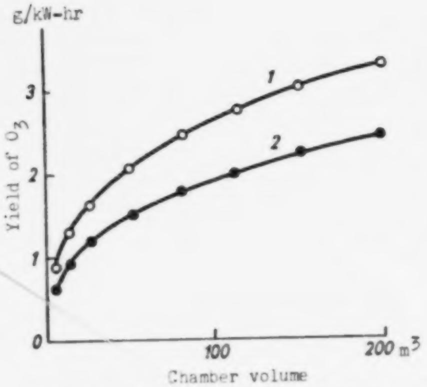

Fig. 3. Yield of ozone in chamber with PRK lamp as a function of chamber volume.
1 - PRK-7 lamp, 2 - PRK-4 lamp

The quantum yield of photochemical oxidation of nitrogen does not exceed 0.03 molecule, and the energy yield is no greater than 0.4 molecule of nitrogen dioxide per 100 eV (7 g/kW-hr). Thus, when PRK type lamps act on pure air, 50-60 times less nitrogen oxides are formed than ozone. The formation of ozone in air containing nitric oxide under the influence of ultraviolet radiation takes place with an energy yield of 460 g/kW-hr, and in pure air containing nitrogen dioxide, 680 g/kW-hr. Thus, nitrogen oxides accelerate the formation of ozone only slightly in the presence of light with wavelength up to 253 nm. When nitrogen dioxide is decomposed at wavelengths up to 395 nm, the process of ozone formation is not limited to the depletion of nitric oxide (process 7), since its concentrations are restored in reaction (8). The formation in (8) of atomic oxygen and also nitric oxide, and the subsequent formation of ozone (reactions 7 and 10) are key factors in bringing about the photochemical reaction in atmospheric air. Since, however, the formation of ozone according to reaction (10) is a much faster process than the oxidation of nitric oxide, the rate-determining step in the photochemical reaction in atmospheric air is the oxidation of nitric oxide. Moreover, in a slow process of nitrogen oxidation, the ozone formed interacts with nitric oxide, so that its steady-state concentration remains insignificant.

In the photochemical formation of ozone in the presence of nitrogen oxides, nitrogen atoms are also formed to a slight degree, particularly in the following reactions:

$$NO^* + O \rightarrow O_2 + N. \qquad (16)$$

$$NO^* + NO \rightarrow NO_2 + N. \qquad (17)$$

$$N_2O + O \rightarrow NO_2 + N. \qquad (18)$$

The chief process of formation of nitrogen atoms is reaction (16), in which the excitation energy of initial atoms does not exceed 1.5 eV, which corresponds to a wavelength of 800 nm. The energy yield of formation of nitrogen atoms does not exceed 0.1 atom per 100 eV, but the reactions between them, organic compounds, and ozone may lead to the formation of highly toxic nitrogen containing oxidants (5). The presence of hydrocarbons leads to the photochemical formation of peroxides and other oxygen-containing compounds that readily give up an oxygen atom, and are thus able to oxidize nitric oxide and form ozone. As a result of the accumulation of perioxides and acceleration of the oxidation of nitric oxide, the energy yield and concentrations of the ozone formed in the presence of hydrocarbons increase by a factor of 3-5, as in the case of the influence of nitrogen oxides. A substantial influence of hydrocarbons on the rate of the photochemical reaction is observed only when the concentration of hydrocarbons exceeds that of nitric oxide by a factor of not less than 2-3.

In a static system, when ultraviolet radiation with a wavelength above 260 nm is used, which cannot induce photochemical reactions in pure air, the kinetic equation for the formation of ozone in air has the form

$$v(O_3) = kI\sqrt{[NO]}\sqrt{[NO] + \sum_{i=1}^{n} \alpha_i C_i}\,. \qquad (19)$$

where $v(O_3)$ is the rate of ozone formation, I is the intensity of ultraviolet radiation, [NO] is the nitric oxide concentration, C_1 is the hydrocarbon concentration, α_i is the ozone-forming efficiency of the hydrocarbon relative to nitric oxide, dependent on the chemical properties of the hydrocarbon, and k is a proportionality coefficient.

To confirm the results obtained, of major interest are measurements made with a DRVE-200 type lamp. A study of the spectral emission characteristic of this lamp showed that the lower boundary of its spectrum begins at 280 nm, and the main fraction of ultraviolet radiation is located in the 295-320 nm range, which is close to the spectrum of natural radiation. It was found that when pure air is irradiated, no ozone and no nitrogen oxides are formed. When air is irradiated in the presence of nitric oxide and hydrocarbons, the concentration of oxidants after 1 hour reaches 3 mg/m^3 for 1-hexene, 1 mg/m^3 for tetrahydrofuran, and 0.7 mg/m^3 for cyclohexane (Fig. 4). The highest ozone-forming efficiency is displayed by unsaturated hydrocarbons.

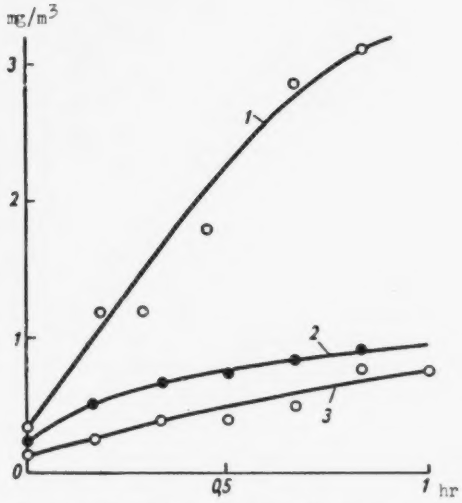

Fig. 4. Concentration of oxidants as a function of irradiation time for DRVE-200 lamp with a chamber volume of 100 l, concentration of nitric oxide of 10 mg/m^3, and hydrocarbon concentration of 35 mg/m^3. 1 - 1-hexene, 2 - tetrahydrofuran, 3 - cyclohexane.

Photochemical reactions may take place with practically any organic pollutants. For this reason, their hygienic importance is particularly great in emissions of low-toxicity compounds in large concentrations. In atmospheric air, the photochemical oxidation of these compounds may lead to the formation of toxic products harmful to public health. As an example, let us cite an experiment with air containing ethyl alcohol (i.e., a weakly toxic compound present in the emissions of the hydrolysis industry). The experiment consisted in studying air on a mass spectrometer before and after a physicochemical action (by using the simulation method under laboratory conditions). The data obtained are shown in Fig. 5 in the form of a dependence of the ionization current (proportional to the concentration) on the time of recording on an automatic potentiometer. As is evident from the figure, before the irradiation, only the ethanol peak was recorded. After the irradiation, the ethanol peak decreased slightly, but peaks of new compounds appeared. The identification of the compound was made by using a magnetic field whose strength was monitored during the recording. The mass spectrograms obtained made it possible to determine the concentrations, which are listed in Table 1.

As is evident from the table, the physicochemical action caused ethyl alcohol to form at least seven new compounds. Of these, three (methanol, acetaldehyde, and formaldehyde) are much more toxic than the initial pollutant, ethanol. As a result, the concentrations of acetaldehyde and formaldehyde formed after the physicochemical action exceeded the MPC approximately 900-fold.

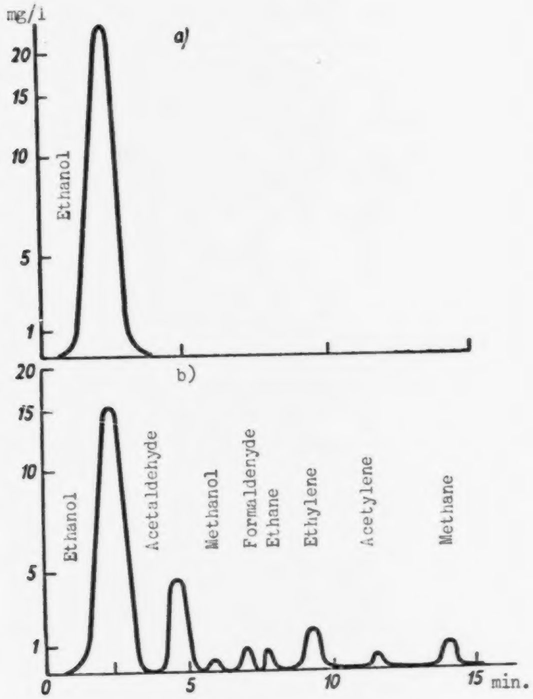

Fig. 5. Mass spectrogram of the products of photochemical reactions of ethyl alcohol in air before (a) and after irradiation (b).

Table 1

Products of Photochemical Reactions of Ethyl Alcohol in Air and Toxicity Factor Before and After Irradiation

Compound	Chemical Formula	Initial Mixture, mg/l	Mixture Obtained, mg/l	MPC For Work Areas	Ratio of Concentrations to MPC
Ethanol	C_2H_5OH	24,2	15,1	1	15,1
Methanol	CH_3OH	—	0,11	0,05	2,2
Acetaldehyde	CH_3CHO	—	4,45	0,005	890
Formaldehyde	CH_2O	—	0,93	0.001	930
Methane	CH_4	—	0,85	—	—
Ethane	C_2H_6	—	0,73	—	—
Acetylene	C_2H_2	—	0,31	—	—
Ethylene	C_2H_4	—	1,65	—	—
Toxicity Factor (relative to MPC)		24,2	1837	—	—

Whereas in the initial mixture the ethanol concentration exceeded the MPC 24-fold, in the mixture obtained after the physicochemical action, the concentrations of the four main toxic substances (formaldehyde, acetaldehyde, ethanol and methanol) exceeded the MPC 1840-fold. Thus, after the action was applied, the toxicity of the mixture formed increased 76-fold.

Since ethyl alcohol is a fairly stable chemical compound with saturated bonds, it may be expected that similar results can be obtained with any other compounds of low toxicity. For compounds with unsaturated bonds, the role of physicochemical factors is obviously much greater.

If one proceeds from the fact that radiation with wavelength of less than 290 nm completely fails to penetrate down to ground level, the following conclusions may be drawn from the above photochemical study. The formation of nitrogen oxides and ozone in pure air is almost completely excluded. The largest amount of ozone is formed by the action of light with wavelength of less than 580 nm in air polluted with nitric oxide (this process is also observed in the wintertime). The chief formation of ozone, resembling atmospheric pollution in character, may take place under the influence of light with a wavelength of less than 395 nm in air polluted with nitrogen oxides. The rate-determining step in the overall process is the oxidation of nitric oxide. The presence of hydrocarbons, mainly unsaturated ones, intensifies the processes of oxidation of nitric oxide and accumulation of ozone, and promotes the formation of nitrogen-containing organic compounds. The reactions between ozone, atomic oxygen and nitrogen, nitrogen oxides, and organic compounds lead to the formation of a group of oxidants that are the components of photochemical smog.

Full-scale observations were made in different cities under different meteorological conditions. The principal results on the concentrations of ozone (or oxidants) are shown in Table 2. It is evident from the table that under the conditions associated with the polluted atmosphere of cities, substantial ozone and oxidant concentrations are observed which considerably surpass the ozone concentrations in pure air. Thus, in the summertime in the center of Moscow, concentrations up to 400 $\mu g/m^3$ were observed, with the ozone concentration proportional to the concentration of nitrogen oxides and to the square root of the concentration of hydrocarbons. The ozone concentration in the wintertime reached 10-200 $\mu g/m^3$. In Baku, the total amount of oxidants at the intersection of Samed Vurgun and Bakikhanov Streets was 70-80 $\mu g/m^3$, and in the area of the Karayev Petroleum Refinery, up to 150 $\mu g/m^3$. In sunny weather, the averaged concentrations were 2.5 times, minimum ones 2.3, and maximum ones 1.9 times as high as in cloudy weather. In southern cities, the average concentrations in cloudy weather were 2 times as high as in Moscow, and in sunny weather, 1.5 times, which is due to more solar radiation. On the basis of individual observations, the maximum concentrations in Moscow are higher than in southern cities, which may be due to a higher content of exhaust gases.

Table 2

Ratio of Average, Maximum and Minimum Values of Ozone Concentration in Sunny and Cloudy Weather for Different Points

Point	Number of Observations	Concentration		
		Av.	Min.	Max.
Moscow:				
Pogodinskaya St.	75	2,3	3,0	2,0
Samotechnaya Pl.	75	4,5	4,0	1,5
Krasnaya Presnya St.	112	2,2	1,9	2,4
Kotel'nicheskaya Quay	44	1,5	2,1	2,0
Baku:				
Vurgun St.	55	2,3	3,0	2,0
Im. 22 Parts'yezda	17	3,0	2,5	1,2
Karayev Petroleum Refinery	30	2,2	2,0	1,9
Yerevan, Markaryan St.	32	1,8	2,0	1,3
Batumi, Petroleum Refinery	45	2,0	2,0	2,3
Average	—	2,5	2,3	1,9

Since one of the most sensitive indices of the occurrence of photochemical reactions is the oxidation of nitric oxide, the dynamics of the ratio of nitric oxide to nitrogen dioxide concentrations under different conditions was investigated. All the emission sources of nitrogen oxides contained over 95% nitric oxide in the total oxides. It is generally assumed that nitric oxide rapidly oxidizes in air as a result of the reaction

$$2NO + O_2 \rightarrow 2NO_2 \, (20).$$

Moreover, the kinetic equation of the oxidation of nitric oxide by oxygen corresponds to a third-order reaction, and the rate of the oxidation reaction is proportional to the square of the nitric oxide concentration. For this reason, reaction (20) has real significance only at concentrations of the order of 1 g/m^3, which are practically never found in atmospheric air. At a concentration of 50 mg/m^3, the oxidation of 50% of nitric oxide takes place in 10 hours. Hence, at a concentration of 5 mg/m^3, such a degree of oxidation will be reached only after 40 days. Thus, reaction (20) practically does not occur under the conditions prevailing in atmospheric air. However, in atmospheric air, the ratio of NO to NO_2 changes continuously as a result of reaction (7).

As an example, Fig. 6 shows typical variations of the ratio of nitric oxide to nitrogen dioxide concentrations with the time of day. For a total content of nitrogen oxides from 20 μg/m^3 to 2.5 mg/m^3, the minimum value of this ratio is 1.8-2.6 in summer and 4.8-8.5 in autumn and winter. On the average, in summer, of the total oxides, NO_2 accounts for 18% in autumn and 9% in winter.* As is evident from the figure, soon after midday, the ratio of nitric oxide to nitrogen dioxide concentrations is rapidly restored, and the concentration of nitric oxide again becomes much larger than that of the dioxide. This is undoubtedly due to the subsequent interaction of nitrogen dioxide with atmospheric moisture, whereby the bulk of the NO_2 deposits with the aerosol, and the remainder is discharged into the air in the form of nitric oxide. Nitric oxide does not react chemically with moisture and is practically insoluble in water.

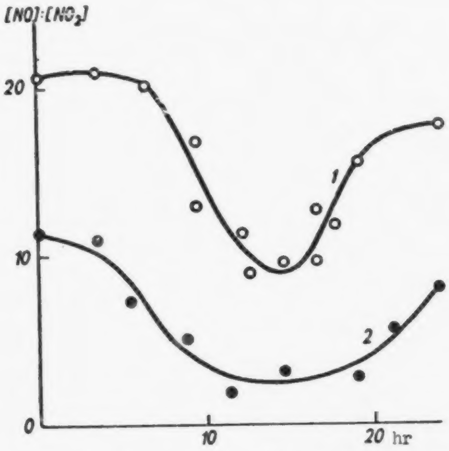

Fig. 6. Ratio of nitric oxide to nitrogen dioxide concentrations as a function of the time of day in autumn (1) and summer (2).

Fig. 7 shows the variations of oxidant concentrations with the time of day, which reveal a distinct relationship to the action of solar radiation. Whereas at 8-9 A.M. the oxidant concentration was 30-60 $\mu g/m^3$, around 12-2 P.M. it rose to 80-170 $\mu g/m^3$. Starting at 3 P.M., the oxidant concentration decreased, and by 7 P.M. amounted to only 10-20 $\mu g/m^3$. The character of this dependence remains unchanged for different geographic zones.

Comparison of the data of Fig. 6 and 7 shows that the decrease in nitric oxide concentration corresponds to an increase in oxidant concentration and vice versa, indicating a close relationship between these two processes.

The unquestionable relationship between the formation of oxidants and the discharge of hydrocarbons is evidenced by the data shown in Fig. 8. As the distance from the local emission source increases, the concentrations of oxidants rapidly decrease. At a distance of over 1 km, the concentrations of oxidants correspond to the background values. In a study of air under conditions of pollution with exhaust gases of motor transport, no relationship was established between the concentration of oxidants and the height of sampling. Thus, at the level of the 12th floor of a highrise building on Kotel'nicheskaya Quay in Moscow, the concentration of oxidants was an average of 20% higher than at ground level, whereas the concentration of oxides was slightly reduced [4].

The concentration of oxidants is affected by other meteorological elements in addition to the solar radiation intensity. The concentrations of ozone and other oxidants decrease with increasing wind velocity, absolute air humidity,

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* [sic.-transl.].

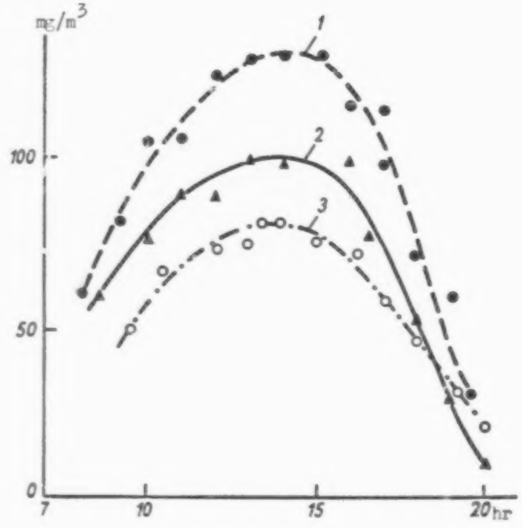

Fig. 7. Concentration of oxidants as a function of the time of day.

1 - Baku, Karayev petroleum refinery, 2 - Moscow, Samotechnaya Plaza, 3 - Baku, street intersection.

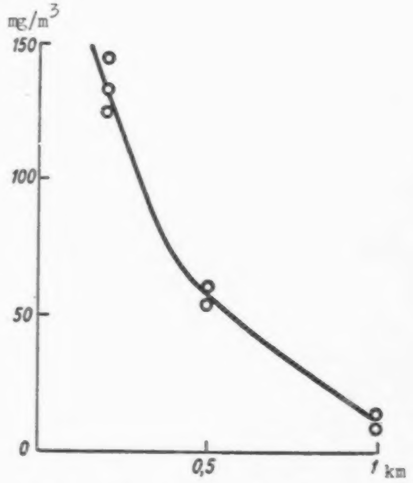

Fig. 8. Average concentrations of oxidants versus distance from local emission source in Baku at the Karayev petroleum refinery.

and rising air temperature. Ozone and other oxidants disappear from air almost completely in summer during precipitation [3]. As the atmospheric pressure rises, the oxidant concentrations increase slightly, but the strongest effect on the increase of oxidant concentrations is caused by temperature inversions associated with the formation of stagnation zones. It follows from the data obtained that the concentrations of oxidants change under the influence of meteorological elements as do ordinary atmospheric pollutants, with the exception of solar radiation and air humidity. As the radiation intensity increases, the concentrations of oxidants rise, and as the humidity increases, they decrease, whereas for ordinary pollutants (for example, sulfur dioxide) the opposite relationships are observed.

Thus, the full-scale studies performed confirm the basic relationships leading to photochemical reactions in atmospheric air. In addition to physicochemical studies, an experimental approach to the study of the biological effect of photochemical reactions is being developed at the present time. Chambers with quartz lamps in which different substances are irradiated have been constructed for this purpose. A study of the biological effect of oxidants established that the odor threshold for ozone (in comparison with pure air) is 18-22 $\mu g/m^3$. The adapted threshold of continually perceived odor (when the subjects are placed in rooms with changing ozone concentration) is 200-250 $\mu g/m^3$. An acute irritation of the mucosa of the eyes is observed at concentrations of 500-700 $\mu g/m^3$. The laboratory and full-scale studies conducted indicate the urgency of the problem of studying photochemical reactions in a polluted atmosphere for cities and industrial centers under the conditions prevailing in the USSR, so that research on this problem will be continued and expanded.

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M. T. DMITRIEV, N. A. KITROSSKIY, V. A. POPOV

MECHANISM OF PHOTOCHEMICAL POLLUTION OF TOWN ATMOSPHERE *

Intensive photochemical reaction in atmospheric layer can go on in the presence of substantial concentrations of nitrogen oxides and 2—3 times as large concentrations of hydrocarbons. In this case, photochemical ozone formation begins under the influence of radiation with the wave length less than 580nm, while in clean air the wave length necessary for this process amounts to 253nm. Ozone formation goes on at oxidation of nitrogen oxide under the influence of light with wave length up to 580nm and at excitation of nitrogen dioxide by radiation with the wave length up to 395nm. Hydrocarbons speed up oxidation of nitrogen oxide which is rate-determining step.

Natural investigations of photochemical reactions in polluted air showed that in sunny weather concentrations of ozone and oxidants are 2—2.5 times as high as in foggy weather. The ratio of nitrogen oxide concentration to that of nitrogen dioxide which amounts to 20—40 at night decreases in the daytime up to 2—5, and then returns again to initial values. For all this, concentrations of ozone and oxidants minimal at night increase sharply in the daytime, especially in sunny weather. Oxidant concentrations decrease significantly with the increase of distance from a local source of hydrocarbon emission. Under the influence of meteorological factors (wind velocity, temperature, atmospheric pressure, lapse rate, precipitation) oxidant concentrations change like common atmospheric pollution.

With the increase of radiation intensity and decrease of air humidity, oxidant concentrations of photochemical origin are growing, while concentrations of common pollution are diminishing.

While studying biological effect of oxidants, it is established that the threshold of comparative smell amounts to 18—22 mcg/m^3, the threshold of continuously perceived smell is 200—250 mcg/m^3, and strong irritation of mucous eyes membrane is observed at 500—700 mcg/m^3.

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* Editor's note: The abstract is presented as given in English with the original Russian article but has been slightly edited.

PROCEDURE FOR DETERMINING THE CONTENT OF TRACE ELEMENTS IN PRECIPITATED WATER

T. N. Zhigalovskaya, R. I. Pervunina, V. V. Yegorov,
E. P. Makhon'ko, and A. I. Shilina (USSR)

From Glavnoe Upravlenie Gidrometeorologicheskoy Sluzhby Pri Sovete Ministrov SSSR. (Chief Administration of the Hydrometeorological Service Under the Council of Ministers of the USSR.) "Meteorologisheskie Aspekty Zagryazneniya Atmosfery". (Meteorological Aspects of Air Pollution.) Sbornik dokladov na mezhdunarodnom simpoziume v Leningrade - Iyul' 1968 g. (Reports delivered at the International Symposium in Leningrad - July 1968.) Pod redaktsiey d-ra fiz.-mat. nauk M. E. Berlyanda. (Edited by Prof. M. E. Berlyand.) Gidrometeorolgicheskoe izdatel'stvo, Leningrad, p. 310-319 (1971). (Hydrometeorological Publishing House, Leningrad, (1971).)

The determination of the content of trace elements in precipitated water and cloud water is of interest in connection with the study of the washing out of atmospheric pollutants by precipitation and the study of transport of cloud masses. Such determinations can give results that are of interest in the geophysical and hygienic sense only when sufficient data are available, and it is therefore necessary to have a method of determination that is as little time-consuming as possible. In addition, because of the low concentration of elements in rain and cloud water, the analytical procedure should be highly sensitive, and the preliminary preparation of samples should be as simple as possible in order to minimize the probability of introducing contaminants.

The object of the present work was to develop a sensitive spectral procedure for the simultaneous determination of the following trace elements in samples of precipitated water: lead, chromium, vanadium, manganese, cadmium, nickel, cobalt, molybdenum, copper, zinc, bismuth, aluminum, and titanium. These components were selected as the most important ones for air hygiene.

In most cases, the content of trace elements in atmospheric precipitation is below the sensitivity of a direct determination. To raise the concentration of the elements studied, preliminary concentration is employed.

1. Selection of the Method of Concentrating Trace Elements in the Analysis of Rain and Cloud Water

At the present time, the following concentration methods are used in analyzing liquid samples for the content of trace elements:

(1) extraction [3, 2];
(2) cation exchange [1, 9];
(3) evaporation [2].

In selecting the concentration method, analysis of samples of rain and cloud water for the content of trace elements should consider the following:

(a) degree of concentration;
(b) probability of introduction of contaminants;
(c) time and labor required when the method is used for mass analyses.

Comparative values of the degree of concentration by means of the above-indicated methods are cited by V. Ya. Yeremenko [3]. Concentration by extraction makes it possible to obtain the highest degree of concentration. However, along with the high degree of concentration, the extraction method also has its drawbacks. One of them is the use of a large number of reagents, which must be thoroughly purified.

Higher grade reagents, i. e., chemically pure ones (c. p.), contain a comparatively large number of impurities amounting to hundredths of a percent, and only in acids (hydrochloric, nitric, sulfuric) not more than 2×10^{-3}% of impurities are allowed [6, 8]. Such amounts of impurities do not affect the results in the analysis of samples with high concentrations of elements.

In the analysis of samples containing trace quantities of elements of the order of 10^{-4} mole/l or less, these reactants must be additionally purified. When a large number of trace elements are determined in the same sample, the extraction method requires a separate isolation of the group of elements by various extracting agents, and this presents a certain difficulty in the application of this method to mass analyses of samples.

Concentration by cation exchange involves a lesser degree of concentration as compared with extraction. However, this method makes it possible to use a smaller number of reactants, mainly, hydrochloric acid of v. p. grade and a cation exchanger. Hence, fewer contaminants are introduced in the course of cation exchange. Nevertheless, when concentrating trace elements by cation exchange, one must consider the results of blank tests. Following the concentration, the background levels of the trace elements may be higher than the ones being determined, since according to our calculations, acid of v. p. grade contains $1\text{-}5 \times 10^{-5}$ mg/ml of impurities. Thus, when 50 ml of 4 N hydrochloric acid is used for elution and an overall 100-fold concentration, as in our case, the method is applicable to trace element concentrations of the order of 1×10^{-3} mg/l in the sample. In this case, the background concentrations of the elements entering the sample with the acid amount to approximately 2×10^{-5} mg/ml. We used this method to determine the trace elements in rain water in cases where the sample volume was 1-2 ℓ. The use of the method loses meaning when it is mecessary to analyze samples less than 1 ℓ in volume. The difficulty of using the method in mass analyses for the content of trace elements lies in the preparation of the cation exchanger.

Evaporation yields the same degree of concentration as cation exchange, but in contrast to the latter, the evaporation method uses only one reagent, hydrochloric acid. In evaporation (especially in glass vessels), the concentrate obtained may be contaminated with additional impurities due to a certain solubility of the glass [7]. For this reason, it is recommended that the evaporation be done in platinum or quartz crucibles. Other preventive measures aimed at achieving an adequate purity should also be employed, namely: the sample shoud be evaporated in portions over a water bath, the crucible containing the sample being covered with an inverted funnel. To decrease the adsorption of the elements on the walls of the vessel used for evaporation, hydrochloric acid of c. p. grade should be used in the amount of 1 ml per 100 ml of sample.

If a sample with a volume of 100 ml is brought down to 1 ml, the amount of acid in the sample following the concentration remains equal to 1 ml. One ml of hydrochloric acid of v. p. grade, which we used, contains about 1 x 10^{-5} mg/ml of each of the elements. Let us postulate that the trace element being determined is present in water in a concentration of about 1 x 10^{-3} mg/l or 1 x 10^{-4} mg/ml, this being one order of magnitude larger than the concentration of the element in 1 ml of acid, and hence, fully admissible. We used the method to analyze samples less than 500 ml in volume. In this case, the errors introduced by the reagents were slight.

In the determination of trace elements in cloud water collected during the study of the washing out of atmospheric aerosols, or in the determination of trace elements in rain collected in fractions, the sample volume may be less than 100 ml. For the latter case, we developed a method of concentration by electrolytic deposition on a carbon electrode. The method consisted in isolating from the sample the elements being determined by depositing them on a carbon electrode 3 mm in diameter serving as the cathode. The pretreated electrode was placed in a beaker containing the sample and connected to the anode compartment by an electric bridge. A platinum wire placed in a beaker containing 0.1 N nitric acid of v. p. grade served as the anode.

The intensity of the spectral lines, proportional to the amount of substance deposited on the electrode, depends on the method of treatment of the electrode, current intensity, acidity of the solution and electrolysis time, as shown in Tables 1, 2, and 3.

Table 1

Deposition Coefficient as a Function of the Normality of Solution (I = 20 mA, electrolysis time t = 2 hours, C = 0.005 mg/l)

Normality of solution, N	Ni	Bi	Al	Pb	Mn	Cr	Fe	Mo	V	Sn	Cd	Ti	Cu	Zn	Co
0,5	—	0,46	1,04	0,22	0,22	—	0,92	—	—	—	—	—	0,87	0,80	—
0,1	0,61	0,75	1,25	0,30	0,9	0,36	1,81	0,15	0,38	0,3	0,08	0,55	2,15	1,35	0,3
0,01	0,5	0,88	1,04	—	0,82	0,24	1,84	0,36	0,42	0,21	0,35	0,78	2,2	1,85	0,45

Table 2

Deposition Coefficient as a Function of Current Intensity
(0.01 N, electrolysis time t = 2 hours, C = 0.005 mg/l)

Current intensity, mA	Zn	Fe	Bi	Al	Ti	Sn	V	Mo	Pb	Cu
40	1,15	1,28	0,57	1,17	0,41	0,21	—	0,20	1,39	2,5
20	0,39	0,63	0,32	0,77	0,31	0,21	0,18	—	0,42	2,20
10	—	0,51	0,28	0,64	—	0,08	0,42	—	0,12	1,68

Table 3

Optical Densities of Spectral Lines in the Electrodeposition of Metals From 10 ml of Solution with 0.0025 mg/l Concentration of Each Metal

Electrolysis time, hr	Fe	Ni	Co	V	Cr	Mn	Sn
1	0,6	0,16	0,22	0,20	0,23	0,35	0,13
2	1,65	0,25	0,40	0,22	0,22	1,74	0,23
3	1,48	0,22	0,25	0,20	0,20	0,78	0,26
Electrolysis of distilled water, 2 hours	1,01	—	—	—	—	—	—
Relative sensitivity, %	$3\cdot10^{-8}$	$2\cdot10^{-8}$	$2\cdot10^{-7}$	$2\cdot10^{-8}$	$2\cdot10^{-8}$	$2\cdot10^{-8}$	$2\cdot10^{-8}$

Electrolysis time, hr	Pb	Zn	Bi	Al	Cd	Cu
1	0,34	0,35	—	0,98	0,28	1,97
2	0,58	1,50	0,15	1,40	0,25	2,80
3	0,88	1,57	0,42	1,32	0,29	2,14
Electrolysis of distilled water, 2 hours	—	0,49	—	0,57	—	1,28
Relative sensitivity, %	$5\cdot10^{-8}$	$3\cdot10^{-8}$	$1\cdot10^{-8}$	$3\cdot10^{-8}$	$2\cdot10^{-7}$	$1\cdot10^{-10}$

The electrolysis conditions were selected on the basis of these experiments. The concentration method described combines the enrichment and deposition of the sample on the electrode. This raises the degree of concentration and shortens the time of analysis by a factor of 3-4. The method was checked on solutions of salts of trace elements in doubly distilled water and on samples of the deposits, and was compared with the evaporation method. During the combustion of a sample deposited on the electrode by electrodeposition, the intensity of the lines of the elements increases as a result of a more uniform coating of the electrode and a stronger bond between the elements and the electrode material. As a result, the sensitivity of the determination of the trace elements increases by one order of magnitude.

On the basis of a check of the applicability of the above-indicated concentration methods to the analysis of rain and cloud water samples for the content of trace elements, it was concluded that the concentration method should be selected to conform to the initial volume of the sample.

The actual volume of a single sample of rain or cloud water, collected at temperate latitudes, may be no larger than 500 ml. For this reason, we carried out the concentration by using an evaporation method that we developed for analyzing rain water for the content of trace elements. In the case of analysis of samples less than 100 ml in volume, the electrolytic concentration method was used, followed by spectral determination.

2. Development of the Method of Spectral Analysis

The method of emission spectral analysis permits the simultaneous determination of a large number of elements in a single sample with a sufficiently high sensitivity of the analysis.

The sensitivity of the spectral analysis depends chiefly on the mode of introduction of the sample into the discharge and on the characteristics of the discharge plasma. For this reason, we conducted preliminary experiments to study the sensitivity of the spectral method as a function of the mode of introduction of the sample into the discharge and of the discharge conditions. In so doing we attempted to make the analysis as little time-consuming as possible, since this is an indispensable requirement in the mass analysis of samples.

Following the preliminary concentration, the samples of rain and cloud water were analyzed spectrally by using the method of three standards. According to the procedure we developed, the samples of rain and cloud water are analyzed without being converted to a dry residue. This facilitates the standardization, makes it possible to increase the accuracy and sensitivity of the method, and eliminates the necessity of grinding and mixing the samples during their preliminary treatment. Thus the probability of introducing contaminants is decreased, and the analysis becomes less time-consuming.

The standard solutions are prepared separately for each of the elements being determined: Pa, Cr, V, Mn, Cd, Ni, Co, Sn, Mo, Cu. Zn, and Ti. The substances we used to prepare standards of c. p. grade are shown in Table 4. The latter also lists weighed samples based on 250 ml of solution with a concentration of 1000 mg/l of the element being determined.

The material from which the laboratory ware and the containers for storing the standard solutions were prepared may also be a source of contaminants and cause a decrease in the concentration of the solutions they contain. In [4], it is shown that much smaller amounts of contaminants enter the liquids from the polyethylene containers in which they are stored than from glass containers. Moreover, after prolonged storage of standard solutions in polyethylene ware, the composition of the solution remains practically unchanged.

In the above-described method of preparation and storage of standards, the concentration of the elements subsequently changes at a slow rate, so that it may be considered constant over a period of six months. In the analyses, mixtures of standard solutions of different concentrations were employed. These mixtures were prepared by the method of successive dilution. Thus, mixtures containing all the elements being determined in concentrations of 25, 10, 5, 2.5, and 1.25 mg/l were prepared from standard mixtures with a concentration of 1000 mg/l. Before the analysis, the standard solutions are poured into clean weighing bottles and diluted four times with doubly distilled water. In diluting the standards, it is necessary to use laboratory ware made of stable "pyrex" grades of glass or quartz.

Table 4

Substances Used for Preparing Standards

Standard, 1000 mg/l	Recommended compound	Amount, mg	Solvent	Remarks
Aluminum	Al metal	250	HCl	Dissolve with heating
Nickel	$Ni(OH)_2$	394	HCl	
Iron	$FeCl_3 \cdot 6H_2O$	1211	HCl	
Cobalt	$Co(OH)_3$	466	HCl	
Zinc	Zn metal	250	HCl	
Tin	Sn metal	250	HCl	
Vanadium	NH_4VO_3	574	HCl	
Cadmium	$Cd(OH)_2$	325	HCl	
Bismuth	Bi metal	250	$HNO_3 + HCl$	Dissolve with heating
Chromium	K_2CrO_4	933	HCl	
Manganese	$MnCl_2 \cdot 4H_2O$	901	HCl	
Copper	Cu metal	250	$HNO_3 + HCl$	Dissolve with heating
Molybdenum	Mo metal	250	HCl	
Lead	$Pb(NO_3)_2$	400	HCl	

To excite the spectra, the samples and standards are introduced into a discharge obtained with a standard arc generator. The electrodes used were of spectroscopically pure carbon, and this made it possible to achieve a sufficiently high excitation temperature and purity of the discharge plasma [5, 10].

In order to study the intensity of the spectral lines as a function of the current intensity, electrode shape, and depth of penetration of the sample into the electrode, special experiments were set up. It was assumed that the location of the sample on the electrode surface makes it possible to increase the accuracy of the analysis, since the vaporization of the sample will be independent of the differences in electrode porosity. We showed that the sensitivity of the analysis could be increased by decreasing the convection currents. To this end, one should select the current intensity and discharge frequency so that the electrodes heat up only on the surface where the sample has deposited. The use of plane electrodes arranged vertically could also result in a decrease of the convection currents.

Experiments on the intensity of spectral lines of the elements as a function of the method of electrode surface treatment and discharge conditions were used to select the conditions of excitation of the spectra. Reproducibility of the analysis is achieved by the absence of mutual influence of the elements. To this end, after the electrodes have dried, a drop of a sodium solution with a sodium ion concentration of 1000 mg/l is deposited on each electrode.

A DG-2 generator is used as the excitation source. The spectra are photographed at a current intensity of 3.5 A and an exposure of 10 sec. Above this intensity, as shown by high-speed motion-picture data, explosive phenomena are observed near the electrode surface. This causes a decrease of sensitivity and increase of random errors.

The evaporation of the samples from the end surfaces of the electrodes and the excitation of the spectra are carried out with vertical electrodes, the distance between the electrodes being 2.5 mm. The spectra of the elements being determined, excited in an alternating-current arc, are obtained by means of a type ISP-28 quartz spectrograph and recorded on aerial photographic film with a sensitivity of 1300 u. GOST-0.85 and a contrast of 2_u. The analytical lines of the elements being determined are photometered on an MF-2 microphotometer. The film background near the photometer lines is used as the internal standard. The concentrations of the elements are determined from the photometric data for the standards and samples.

We found that when this procedure is used and the concentration of one element exceeds the others by a factor of 10^3, the line intensities of the other elements remain constant within the measurement errors. Thus, there is virtually no mutual influence between the elements.

Table 5

Intervals of Concentrations of the Elements Being Determined in Rain Water

Element	Concentrations, μg/l	Element	Concentrations, μg/l
Sn	1,0—60	Al	0,3—30
V	1,0—60	Co	0,4—60
Fe	0,05—60	Pb	2,0—60
Ni	2,0—60	Zn	0,2—60
Mn	1,0—60	Cu	0,12—6
Cr	3,0—60	Cd	2,0—60
Bi	0,3—60	Mo	0,5—30

Under the conditions of the experiments described, the mutual influence is mainly due to the plasma temperature, concentration of thermal electrons, and distribution of matter over the discharge radius.

Because of the high sodium content (20 μg) relative to the amount of sample (0.1 μg), the sample composition has no influence on the above-enumerated factors, since the discharge temperature is determined in this case chiefly by the ionization potential of the sodium atoms, and this accounts for the good reproducibility of the spectral line intensity for different contents of the element present in the sample in higher concentration.

The limits of applicability of the simultaneous spectral determination of the trace elements present in the sample in different proportions are shown in Table 5.

The relative mean square error amounts to not more than 20% for a sensitivity of the method of 10^{-9}-10^{-10} g of the substance on the electrode (Table 6).

Table 6

Sensitivity of Spectral Analysis in the Determination of Trace Elements

	Fe	Ni	Co	V	Cr	Mn	Sn	Pb
Authors' data	$5 \cdot 10^{-9}$	$1 \cdot 10^{-8}$	$1 \cdot 10^{-8}$	$9 \cdot 10^{-9}$	$2 \cdot 10^{-8}$	$4 \cdot 10^{-9}$	$7 \cdot 10^{-9}$	$2 \cdot 10^{-9}$
Literature data	$5 \cdot 10^{-8}$	$3 \cdot 10^{-8}$	$5 \cdot 10^{-9}$	$4 \cdot 10^{-9}$	$4 \cdot 10^{-9}$	$1 \cdot 10^{-9}$	$2 \cdot 10^{-8}$	—

	Zn	Bi	Al	Cd	Cu	Mo	Ag
Authors' data	$1 \cdot 10^{-8}$	$5 \cdot 10^{-9}$	$3 \cdot 10^{-9}$	$2 \cdot 10^{-8}$	$5 \cdot 10^{-11}$	$3 \cdot 10^{-9}$	$2 \cdot 10^{-11}$
Literature data	$4 \cdot 10^{-8}$	$2 \cdot 10^{-8}$	$1 \cdot 10^{-8}$	$1 \cdot 10^{-7}$	—	—	—

Table 7 gives an example of the use of the spectral procedure described in the analysis of rain water for the content of trace elements. Similar data on the lead content of rain water samples are given in [11].

Table 7

Content of Certain Trace Elements in Rain Water (mg/l), 1967.

Date	Cr	Mn	Ni	Al	Sn	Pb
24/IX	0,02	0,03	0,02	0,03	0,01	0,01
26/IX	0,01	0,03	0,02	0,08	0,01	0,04
28/IX	0,02	0,01	0,02	0,05	None	0,03
30/IX	0,02	0,01	0,06	0,01	0,01	0,1
2/X	0,01	0,01	0,01	0,03	0,01	0,01

Conclusions

1. To determine the content of trace elements in rain water, the concentration method must be selected in accordance with the volume of the sample.

2. Concentration by cation exchange should be used when the sample volume is over 1 l.

3. For a sample volume of less than 500 ml but more than 100 ml, the concentration should be carried out by evaporation.

4. For a sample volume of less than 100 ml, we worked out a method of concentration by electrodeposition, which shortens the analysis time by a factor of 3-4.

5. The extraction method is not convenient for analyses of rain and cloud water.

6. The proposed method of spectrochemical determination of the concentration of trace elements makes it possible to perform the analysis in the concentration range from 1 to 100 μg/l with an average relative error of not more than 20%. There is virtually no mutual influence of the elements in the proposed procedure.

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T. N. GIGALOVSKAYA, R. I. PERVUNINA, V. V. EGOROV, E. P. MAHONKO, A. I. SHILINA

THE ESTIMATION OF MICROELEMENT CONTENT IN THE WATER OF PRECIPITATION*

Sensitive spectral method of simultaneous content determination of some important, in hygienic respect, microelements, such as lead, chromium, vanadium, manganese, cadmium, nickel, cobalt, molybdenum, copper, zinc, bismuth, aluminium, titanium, in the samples of precipitation and cloud water is described.

As the content of microelements in atmospheric water is usually too low to be evaluated in a direct way, the possible means of sample enrichment are discussed in detail and the area of their employment is evaluated. While analyzing liquid samples for concentrating microelements determined in them, methods of extraction, evaporation, and method employing an ion-exchange resin are mainly used. The examination and analysis of different means of microelement concentrating in the samples of rain and cloud water showed that the concentrating method must be chosen in accordance with initial sample volume. The value being more than one litre, it is necessary to use ion-exchange resin method; evaporation method, worked out for atmospheric waters, is recommended when the sample volume is less than 500ml, but more than 100ml. For sample volume less than 100ml, a specially elaborate method of concentrating by electrolytic precipitation is proposed which consists of isolation of investigated microelements from samples on carbon electrode serving as a cathode. The extraction method is inconvenient for atmospheric water analysis.

According to the method developed by the authors atmospheric water samples are analyzed spectrally without their conversion into dry residue on three standard methods. As an excitation source the alternating-current generator is used. The spectra of determined elements excited in alternating-current arc are received with the help of quartz spectrograph and are registered on high-sensitive aerofilm. Analytical curves of determined element content are evaluated on microphotometer.

In this paper the means of putting sample on electrode and introducing it into the charge are described and applicability of simultaneous definition of spectral microelements contained in the sample are evaluated.

It is shown that the proposed method of chemico-spectral determination of microelement concentration allows to carry out the analysis in concentration range from 1 to 100mcg/l with the mean relative error not more than 20%. The mutual effect of the elements is practically absent.

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* Editor's note: The abstract is presented as given in English with the original Russian article, but has been slightly edited.

CONTENT OF HEAVY METALS IN THE AIR OF CERTAIN REGIONS OF THE USSR

T. N. Zhigalovskaya, V. V. Yegorov, S. G. Malakhov,
A. I. Shilina, and Yu. P. Krasnopevtsev (USSR)

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Heavy metals entering into the composition of the earth's atmosphere, in particular, Pb, Cr, V, Mn, Cd, Ni, Co, Sn, Mo, Cu, Zn, Al and Ti, are classified as trace elements, since they are present in air samples in amounts of 10^{-2}-10^{-12}% [2].

Despite the negligibly small content of trace elemtns in living and nonliving nature, they play an extremely important part in biological processes. The atoms of trace elements enter into the composition of organic molecules, and also into the composition of the internal medium of the organism, ensuring the regulation of its most important processes [6]. A disturbance of the equilibrium of trace elements in the organism leads to pathological deviations from its normal activity. Such disturbances may take place in the case of excess (or deficiency) of individual elements in the environment, despite the ability of higher organisms to extract the needed elements from the environment in the necessary amounts. When they enter the organisms in excess, some of the trace elements (mainly lead) may have a toxic effect. In this respect, it is important to have accurate data on the content of trace elements entering the atmosphere as a result of the activity of industrial enterprises.

The study of the behavior of trace elements in the atmosphere also has important meteorological aspects. They include primarily the study of the laws of propagation of trace elements in the atmosphere from pollution sources, and attempts to develop methods of forecasting the levels of their concentrations in atmospheric air. The reverse problem is also possible, i.e., the use of trace elements as tracers in the study of certain meteorological phenomena. Information on the content of trace elements in the atmosphere may aid in the explanation of the chemical nature of condensation nuclei and their behavior in the atmosphere. Some trace elements may also be useful in many geophysical studies. Thus, their presence in the high layers of the atmosphere and also in the surface layer of air in areas distant from industrial plants (for example, above the ocean, at the poles) permits an evaluation of the chemical composition of aerosols of cosmic origin [7].

All of the above indicates the importance of studying the distribution of trace elements in the environment and provides the basis for its treatment as a medical-biological, hygienic, and geophysical problem.

However, in determining trace elements it is necessary to deal with some major difficulties involved in the measurements of trace amounts of the elements being analyzed. Such measurements are possible only when very delicate methods of analysis are used. This is probably why only a few works are known thus far that are devoted to the study of the content of trace elements, particularly heavy metals, in atmospheric air [9, 10].

Heavy metals in the atmosphere enter into the composition of solid aerosol particles. In accordance with literature data, above industrial areas [8, 9, 10] the variation range of the concentration of solid aerosol particles in atmospheric air extends from 100 $\mu g/m^3$ in areas of light pollution to 4000 $\mu g/m^3$ in London during a heavy smog. The average values at the center of vast industrial areas and cities such as Los Angeles and Cincinnati vary from 200 to 800 $\mu g/m^3$, and in small towns the average concentrations range from 100 to 200 $\mu g/m^3$.

The object of the present study was to obtain preliminary data on the content of certain trace elements entering the atmosphere as a result of human activity in various regions of the Soviet Union. To this end, the contents of lead, chromium, vanadium, manganese, cadmium, nickel, tin, molybdenum, copper and zinc were determined in samples of atmospheric dust collected in 1965-67.

The data given below constitute the first correlation of the material we obtained.

The samples were collected by means of filtering ventilation equipment consisting of a medium-pressure centrifugal fan at the inlet connection of which was mounted a filter holder with filtering material of brand FPP 0.5 m^2 in area. From the known velocity of the drawn air, time of operation of the device, and area of the filter, the volume of air which passed through the device was calculated. The devices operated around the clock for one month. The filters were replaced once a day. After separating the gauze layer, the filters collected during a month at each point were ashed at 500°C. Mean monthly samples were thus obtained. The weight of the ash of clean filters amounted to an average of 250 mg per month. The weight of the dust collected during a month frequently amounted to several tens of grams. For this reason, our calculations neglected the weight of the ash of clean filters as compared with the weight of the sample. The content of trace elements in the clean filters varies over a wide range, but is always two orders of magnitude less than their content in the ash of filters containing a dust sample. This made it possible to neglect the content of trace elements in the clean filters.

Analysis of atmospheric dust samples for the content of the above-indicated trace elements was carried out by using the method of emission spectroscopy [3].

The ashed filters containing the sample are burned in an alternating-current arc. The radiation of excited atoms of the sample is decomposed into a spectrum by means of an ISP-28 spectrograph. The spectrograph is recorded on aerial photographic film with a sensitivity of 1200 u. GOST 0.85. After photometering the obtained spectra on an MF-4 microphotometer, the content of each of the elements is determined. The method we used is characterized by a sensitivity of 1×10^{-10}-1×10^{-8} g on the electrode depending on the element being determined. The relative mean square error of the method is ±20% of the content being determined.

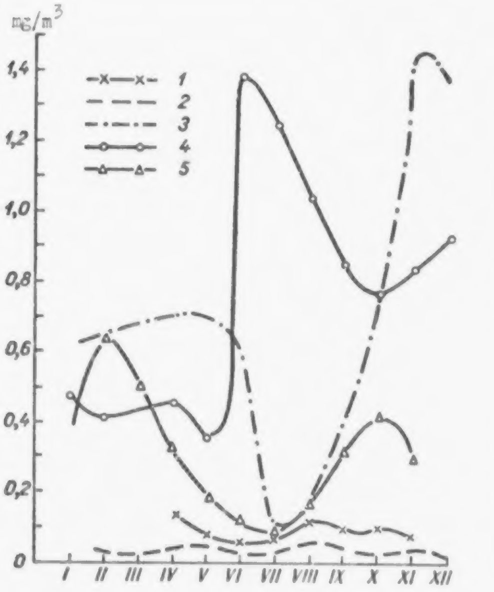

Fig. 1. Total concentrations of trace elements in the air of certain regions of the Soviet Union.

1 - maritime regions of the south, 2 - maritime regions of the north, 3 - maritime regions of the east, 4 - high-mountain regions of Pamir, 5 - regions of industrial cities

The concentration of trace elements in air depends on a set of complex meteorological factors, intensity and periodicity of action of pollution sources, and nature of the terrain. The content of trace elements in the atmospheric surface layer of a specific region may have a seasonal variation depending on the state of atmospheric stability, wind direction and velocity, and degree of washing out by precipitation.

Fig. 1 shows the variation of the total concentration of trace elements which we determined over the course of a year in different regions of the Soviet Union. As follows from the graph, in the air of the maritime regions of the south (curve 1) and north (curve 2) of the country, the content of trace elements increases slightly during the summer months, and remains practically constant the rest of the year. In the maritime region in the east of the country (curve 3), the concentration of trace elements in atmospheric air is much greater than over the northern and southern coasts of the country. The content of trace elements decreases sharply only during the summer months. This is probably due to the direction of the prevailing winds, which in the summer months have a preferred direction from the sea to dry land in these regions [5]. The high-mountain region of Pamir (curve 4) is characterized by an increase in the concentration of trace elements during the summer months, which may be explained by the absence of precipitation and gentle winds during that period, with a consequent weak vertical exchange in the atmosphere [5]. A marked decrease in the concentration of trace elements in these regions was observed in May for both the total concentration and the content of individual trace elements. This is due to the washing out of aerosols from the atmosphere by spring rains [5].

A correlation of the content of trace elements with the annual variation of the amount of precipitation for the high-mountain region of Pamir is shown in Fig. 2b. In the atmospheric air near industrial cities (curve 5, Fig. 1), an approximately 5-fold reduction of the concentration of all the elements took place during the summer months. This correlates satisfactorily with the annual variation of monthly precipitation for the given region, attesting to the washing out of trace elements by atmospheric precipitation (Fig. 2a).

A characteristic element that is continually present in the atmosphere of industrial cities is lead, which enters the atmosphere primarily during the combustion of fuel (for example, gasoline, coal, kerosene, etc.). At the same time, the lead content of atmospheric air is strictly standardized because of the high toxicity of its compounds. It is of interest, therefore, to examine data on seasonal changes of lead in the atmosphere in the vicinity of industrial cities of the country during the period from 1965 to 1967, presented in Fig. 3 (curves 4, 5). For comparison, the same figure shows curves of seasonal variations of the lead content in the atmosphere of maritime and high-mountain regions of the country (curves 1, 2 and 3 respectively). The lowest lead concentrations in atmospheric air were observed in the maritime cities of the south and north of the country. However, in the maritime region of the east of the country, the lead concentration is slightly higher and varies little during the first half of the year, but grows considerably in September-November. It should be noted that in the vicinity of industrial cities (Fig. 3, curves 4 and 5), the total lead content in 1967 was lower than in 1965, i.e., by a factor of 1.5-2. However, in both 1965 and 1966-67, winter concentration maxima were observed. Similar seasonal variations are observed in the surface air in the case of natural radioactive isotopes, for example, radon [4]. A constant source of radon is the earth's surface. The cause of the observed winter maximum in both cases is a weak vertical mixing of air in the lower atmospheric layer in winter, when frequent surface temperature inversions are typical.

For the purpose of sanitary control, it is necessary to have information on the magnitude of the absolute concentration of individual trace elements (for example, lead, chromium, vanadium, etc.) and the total content of dust in atmospheric air. The mean annual values of the concentration of individual trace elements which we obtained for the continental and maritime regions of the Soviet Union are shown in Table 1. For comparison, the table also gives reported data on the content of certain trace elements in the air of industrial cities and suburban areas of the U.S.A. [8, 9, 10], Bulgaria [1], and also data that we obtained from an analysis of aerosol samples above the ocean at 0° latitude and 180° longitude.

Data on the content of chromium, cadmium, nickel, molybdenum, and cobalt are lacking in the literature and are given here for the first time.

We should note the very high level of lead concentration in the air of industrial cities of the U.S.A., more than 40 times as high as the lead

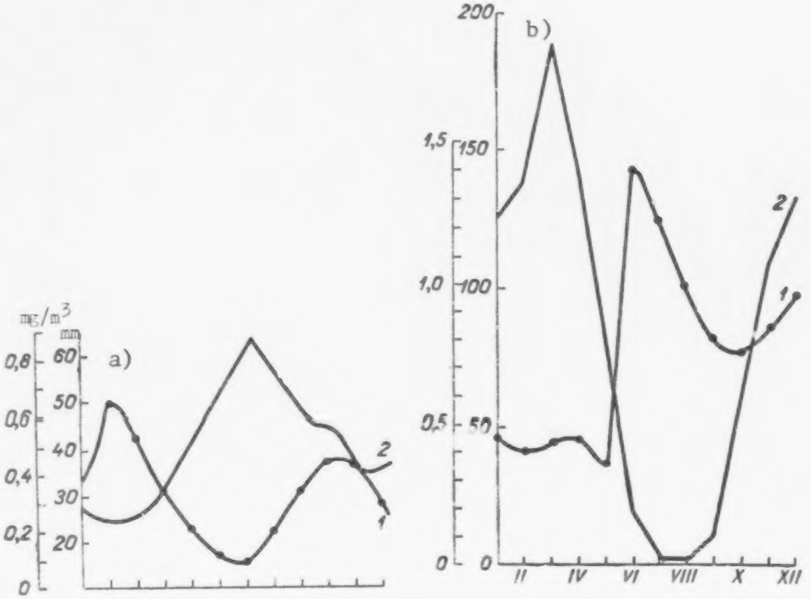

Fig. 2. Annual variation of the amount of precipitation (2) and content of trace elements (1) in the region of an industrial city (a) and in Pamir (b).

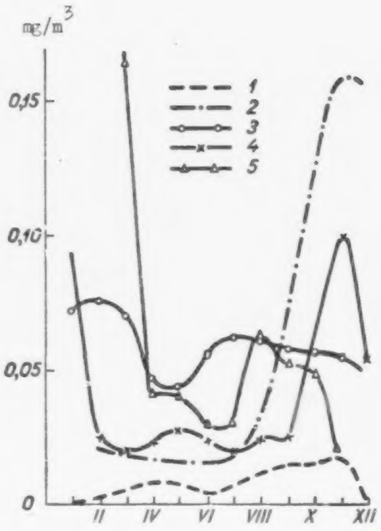

Fig. 3. Annual variation of Pb concentration in the surface air layer for different regions of the USSR.

Table 1

Mean Values of Dust Concentration and Certain Trace Elements in the Surface Atmospheric Layer ($\mu g/m^3$).

Regions	Year	Elements										Dust content	Total content of trace elements	Relative content of trace elements in dust
		Pb	Cr	V	Mn	Cd	Ni	Sn	Mo	Cu	Zn			
Industrial cities of the USSR	1967	0,0301	0,0144	0,0166	0,031	0,0195	0,0211	0,0083	0,0184	0,096	0,040	40	0,2954	0,0074
Maritime cities of the USSR	1967	0,0023	0,008	0,0085	0,015	0,0045	0,017	0,0045	0,0044	0,014	0,047	46	0,1252	0,0027
Industrial cities of the U.S.A. [5, 6]	1956	1,34	—	0,019	0,154	—	—	0,025	—	0,19	—	145	1,728	0,0119
Industrial cities of Bulgaria [9]	1960	1,0–0,5	—	—	0,45–0,20	—	—	—	—	—	—	—	—	—
Suburban areas of the U.S.A.[5, 6]	1956	0,30	—	0,002	0,04	—	—	0,01	—	0,01	—	86	0,362	0,0042
Ocean (180° l, 0° lat)	1966	0,004	0,0008	0,0005	0,0009	—	0,0015	0,002	0,0004	0,016	—	8,7	0,0109	0,0012

concentration in the industrial regions of the USSR. The content of the remaining elements in the industrial regions of the Soviet Union is 2-3 times less than in the industrial cities of the U.S.A. In the maritime regions of this country, the content of most elements is one order of magnitude less than in industrial cities inside the continent. In samples taken above the ocean, the concentrations of all the elements with the exception of lead, tin, and copper are approximately one order of magnitude less than the concentration of these elements in the atmosphere of maritime cities. The higher content of lead, tin, and copper in samples taken over the ocean is probably due to contamination of the samples during their collection on the ship. This applies particularly to the lead content. The lead content was determined in about 40 samples taken on a ship during a cruise in the winter of 1966. In other samples, the lead concentration ranges from 5×10^{-3} to 3×10^{-1} μg/m^3, this being due to pollutants discharged by the stack of the ship and falling on the filter.

In addition, Table 1 lists data on the content of dust, total content of trace elements and ratio of the sum of the trace elements to the total content of dust, i.e., the concentration of the indicated trace elements in dust. The dust concentration in the atmosphere of maritime cities of the USSR is higher than that in the air of industrial cities in the center of the country. The difference in concentrations is close to the dust concentration above the ocean. Thus, the increase in dust concentration above the maritime cities as compared with cities of the continental regions of the country should probably be attributed to marine salts.

The dust concentration above industrial cities of the U.S.A. is approximately 3.5 times the corresponding values for this country and close to the maximum permissible dust norms in the USSR. However, the concentrations of trace elements in dust in 1 m^3 of air were found to be similar in both cases. The concentrations of trace elements in dust in 1 m^3 of surface air of maritime regions of the USSR and suburban areas of the U.S.A. were also similar. For atmospheric dust above the ocean, the ratio of total concentrations of trace elements to the concentration of dust is approximately one order of magnitude less than the corresponding ratio for the air of industrial cities of the continent. Hence, the concentration of trace elements in the dust of the surface atmospheric layer is a measure of the degree of pollution of air with aerosols of industrial origin.

As a result of the above study it may be stated that at the present time, no excess over the established sanitary norms was observed in the average content of trace elements and in their maximum content in the air of the regions of the USSR that we studied. The variation of the concentration of trace elements for continental regions correlates with the amount of precipitation, attesting to the presence of processes of self-cleaning of the atmosphere as a result of washing out. However, because of the growth of industrial cities, the concentration of dust and other polluting ingredients including trace elements may increase in the atmosphere of the globe. From this standpoint, there is need for organizing a regular checking of the content of these impurities in the air of industrial regions, and also regions distant from the pollution sources, in order to determine the variations of the background on a global scale. In this connection, it is of interest to collect air samples over the ocean and at high altitudes.

The preliminary data on the content of trace elements in the surface atmospheric layer, which are presented here, are also of interest in connection with the study of the chemical composition of the atmosphere and processes of atmospheric self-cleaning, which remove dangerous impurities. The table and graphs were compiled on the basis of an analysis of 180 mean monthly samples collected at different points of the Soviet Union in 1965-67, with simultaneous determination of 11 elements in each sample. This volume of work was carried out during a comparatively short period of time, thanks to the use of a semi-rapid spectral method of analysis of adequate sensitivity that we developed. In the future, it is intended to carry out the sampling and analysis of the samples systematically at a large number of points to determine their content of lead, chromium, vanadium, manganese, cadmium, nickel, cobalt, tin, aluminum, titanium, molybdenum, copper, zinc, and iron, which hygienically and geophysically are the most important elements. For comparison, it is intended to conduct analysis of samples taken above the ocean and in the polar regions.

On the basis of the results obtained and of the experimental measurements, particular attention should be directed to the necessity of observing certain operational requirements in collecting and analyzing samples of atmospheric aerosols for the content of trace elements.

In collecting aerosol samples on ships and in polar regions, it is necessary to select the sampling site so as to exclude the possibility of contamination of the sample during the operation of internal combustion engines and by other sources. In analyzing samples for the content of aluminum and iron, contact between the filters and the metal parts of the filter holder is inadmissible.

Clean conditions must be provided during ashing of the filters, and to this end the ashing should be carried out in closed platinum or quartz crucibles.

To exclude a background contaminated with trace elements present in the ash of clean filters, it is necessary to ash and analyze the clean filters of each batch.

When the sampling is conducted in slightly polluted regions, an impinger can be used, and its working surfaces should be coated with polyethylene film.

When the samples are collected on filters in slightly polluted regions, for example, above the ocean or at high altitudes, the content of trace elements in the sample is comparable to their content in a clean filter. In order that the content of trace elements in the sample be above the background produced by the filter, it is necessary to draw a large volume of air through the filter and to know the exact ash content of the clean filter and the content of trace elements in this ash. The weight of the ash must be measured to within 0.1 mg. Data thus obtained may be useful in evaluating the pollution of the lower layers of the earth's atmosphere.

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T. N. GIGALOVSKAYA, V. V. EGOROV, S. G. MALAKHOV,
A. J. SHILINA, Yu. V. KRASNOPEVZEV

THE CONTENT OF HEAVY METALS IN THE AIR OF SOME REGIONS OF THE USSR*

Data on content of lead, chromium, vanadium, manganese, cadmium, nickel, tin, molybdenum, copper, and zinc which get into the atmosphere as a result of human activity are given. These microelements were determined in samples of atmospheric dust collected in different regions of the Soviet Union in 1965—67. The sampling was

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* Editor's note: The abstract is presented as given in English with the original Russian article but has been slightly edited.

carried out with the help of filter-ventilation installation on Petryanov filter with area of $0.5m^2$ which was changed once a day. The filters employed during each month were burned down and ash residue received in such a way was analyzed with the help of emission spectroscopy method developed by the authors. This method is characterized by sensitivity of 1.10^{-10}—1.10^{-8} g on electrode in dependence on determined element. Relative mean-square error of the method amounts to 20% of evaluated content.

The values of total microelement content in the air are given for some regions of the USSR during the whole year. Marked influence of meteorological factors (in particular, wind direction and precipitation) on concentration value is noted. Microelement content in the air closely correlates with annual course of monthly precipitation amount which indicates on significant washing-out action of precipitation. Lead content in the atmosphere is separately considered the presence of lead is due mainly to burning down the fuel. The highest lead concentrations of order of 0.16 mcg/m^3 are discovered near industrial towns such as Kuibyshev, Semipalatinsk, Novosibirsk in spring and in the neighbourhood of Magadan in autumn. Lead content amounts to less than 0.05 mcg/m^3 in the atmosphere of seaside regions of the country and to 0.05—0.08 mcg/m^3 in highland regions.

The values of dust content and absolute microelement concentrations in the air for continental and seaside regions of the Soviet Union and also the data of aerosol content over the ocean at the latitude of 0° and longitude of 180° in 1966 are compared with literature data on microelement content in the air of industrial cities and country regions of the U. S. A. and Bulgaria.

Fulfilled investigations show that at present mean and maximum concentrations of determined microelements in the air do not exceed stated sanitary norms in the examined regions of the USSR. However, in connection with growth of industrial towns it is necessary to carry out regular control of microelement content for evaluation of background variation in global scale near industrial areas and also in the regions removed from pollution sources.

ON THE DESIGN OF A MEASURING NETWORK FOR AIR POLLUTION IN THE GERMAN DEMOCRATIC REPUBLIC*

W. Warmbt GDR (German Democratic Republic)

From Glavnoe Upravlenie Gidrometeorologicheskoy Sluzhby Pri Sovete Ministrov SSSR. (Chief Administration of the Hydrometeorological Service Under the Council of Ministers of the USSR.) "Meteorologisheskie Aspekty Zagryazneniya Atmosfery". (Meteorological Aspects of Air Pollution.) Sbornik dokladov na mezhdunarodnom simpoziume v Leningrade - Iyul' 1968 g. (Reports delivered at the International Symposium in Leningrad - July 1968.) Pod redaktsiey d-ra fiz.-mat. nauk M. E. Berlyanda. (Edited by Prof. M. E. Berlyand.) Gidrometeorolgicheskoe izdatel'stvo, Leningrad, p. 330-336, (1971). (Hydrometeorological Publishing House, Leningrad, (1971).)

1. INTRODUCTION

With the increase in industrialization and of transportation, and with the density of population, pollution of the air with gaseous and solid trace materials has become a real problem of our technological age. The need for solution is pressing. In view of this situation, all the questions and measures to be applied to the problem of air pollution in the German Democratic Republic [GDR] have been coordinated centrally in the Commission for Maintenance of Air Purity. The work of the commission was expressed in the Order for Maintenance of Air Purity published by the Council of Ministers of the GDR in 1966. In this order, the Meteorological Service is assigned the mission of measuring the basic load of air pollutants, particularly the dust and SO_2 concentrations, in the various climatic regions of the GDR. On the other hand, the air pollutants in the major industrial and population points, as well as in working places, are measured by the Hygiene Institutions. Some large factories have begun further to set up their own measuring networks for dust and SO_2. In the design of a measuring network for air pollution, the Meteorological Service is also observing the recommendations of the Aerology Commission of the WMO (CAe - IV/Doc. 11, p. 7), according to which the purpose of the National Meteorological Service is to establish air pollution measuring networks in non-metropolitan areas in order to measure the so-called background concentration.

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* Editor's note: This report was originally presented in German. Its English translation has been supplied by APTIC.

2. ORGANIZATION

The measuring network of the Meteorological Service, German Democratic Republic, will encompass 13 synoptic stations set up in different climatic regions (central mountains, flat land, Baltic Sea Coast). For comparative reasons, stations will also be placed in regions of differing population density in the network.

So as to obtain the most complete survey possible of the trace material content of the atmosphere, the measuring network also includes stations at which atmospheric trace materials and even the ozone near the ground and the radioactivity of the air and precipitation are already being measured. Simultaneous measurements of various trace materials are important not only for a meteorological evaluation of the experimental results, but also for determining the complex biological action of air pollutants.

The Meteorological Service has been performing ground-level ozone measurements in a 6-station network since 1952, and radioactivity measurements in a 9-station network since 1956. Figure 1 shows the distribution of the stations in the air pollution measurement network.

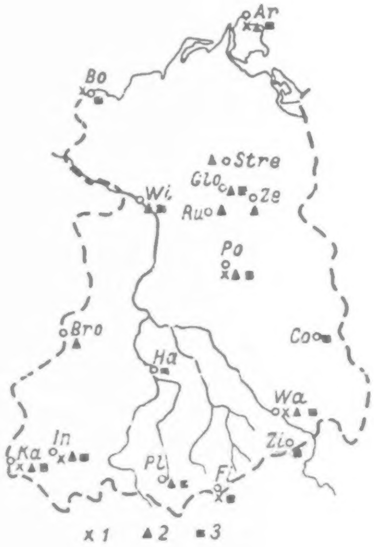

Figure 1. Distribution of stations in the air pollution measuring network.
1. Ozone. 2. Radioactivity.
3. Dust, SO_2.

The Wahnsdorf Meteorological Observatory is the administrative institute for scientific processing of the measurements, as well as for the construction and maintenance of the measuring network. It is also concerned with testing measuring methods and measuring equipment for air pollution research and with problems of measuring network planning. Problems of trace material diffusion are worked on by the Technical Meteorology Group of the Meteorological Service in Potsdam.

3. MEASURING METHODS AND EQUIPMENT

Since 1965, the dust content of the air has been determined in connection with measurements of the radioactivity of the air. The air is drawn through a membrane filter which is as hydrophobic as possible. Czechoslovakian-made filters are used in the net (Type RUFS Synpor 2, Diameter 35 mm, mean pore diameter 2.5 μm). The filter is protected against precipitation and sedimentation of large dust by a small roof. The intake height is 2 m. About 60 m^3 air is drawn through the filter in 24 hours. The filter traps only the fine or suspended dust with particle radii less than 20 μm. The filters are weighed with a sensitive spiral spring balance before and after air sampling. The weight difference and the air volume give the dust content in mg/m^3.

The SO_2 content is measured by the pararosaniline method of West and Gaeke. Measurements are made at the hours of 0, 6, 12, and 18 Central European Time. Samples are taken for a period of 1 hour, or for 30 minutes in more strongly polluted areas. The air flow drawn in is 50 liters per hour. Each station has a spectrophotometer (Spekol) made by the Carl Zeiss Peoples Factory at Jena for colorimetric measurements. The West and Gaeke measuring method has been made obligatory in the RGM* area for SO_2 measurements because it is specific for SO_2. The toxicity of the 0.1 M solution is a disadvantage of this method, as it contains 27.2 g mercuric chloride ($HgCl_2$) per liter. We were able to reduce the amount of poison and perform measurements with a 0.01 M solution without injuring the sensitivity and reproducibility of the measurements. Because of the GDR Poison Law, however, the observer responsible for making the SO_2 measurements at the station must pass a poison examination in order to get a partial poison license.

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*Translator's Note: Unknown geographical area.

4. PLANNING OF THE MEASURING NETWORK

In the performance of network SO_2 measurements, it is important to make the choice of measuring sites so that the measurements are representative for the area in question. We have studied this question in the framework of parallel measurements of SO_2 concentration in the regions of Dresden and Radebeul. We recorded the SO_2 concentration with two Novak analyzers (CSSR). One of these devices was at the Wahnsdorf Meteorological Observatory and the second was in Radebeul, 1.4 km away from the observatory. Similar measurements have recently been made between Wahnsdorf and a measuring site in the city region of Dresden 10 km from the Observatory. There is also an altitude difference of 120 m between Wahnsdorf and the measuring sites in Radebeul and Dresden. We found that, on the average, there were neither large differences in the averages nor in the daily course between these measuring sites. From this, it follows that local differences in the SO_2 content are slight for measuring sites which, as in our example, are within the range of influence of large area sources, as the lower layers of the atmosphere are well mixed horizontally and vertically with SO_2.

Sampling problems appeared only at the measuring site at the Grosser Inselsberg mountain station at 910 m altitude in the Thüringer Forest. The station is in a two-story building complex which also contains the central heating system. With low background concentrations, differences of 10 to 20 times the unperturbed measurement appear on the windward and lee sides of the building, depending on the wind direction. As high wind speeds occur more frequently at this altitude than on flat land, turbulence in the lee of the building is at times particularly pronounced, so that exhaust gas fumes can arrive at the intake opening, depending on the position of the intake and the wind direction. In order to eliminate this effect, the intake position was placed at the tenth level of the television tower.

Another important question of measuring network planning concerns the establishment of the measuring times. Although use of recording equipment for SO_2 measurement is planned, it was studied whether differences appear if averages, frequency distributions, and immission characteristics are calculated from a selection of measuring times or from 24 half-hour measurements. We established that measurements of SO_2 content at the times of 0, 6, 12, and 18 hours Central European Time are sufficient for comprehensive characterization of the air pollution situation, because there were no differences from the statistical measurement coefficients calculated from 24 half-hour measurements.

5. EVALUATION OF THE MEASUREMENTS

It is planned that after 1 January 1969, all the measurements of trace materials will be converted to punched cards and evaluated centrally at a computer institute. In the future, measurements from the hygiene and industrial measuring networks are also to be processed in the same form. Finally, the results of simultaneous SO_2 and dust content measurements will be reported.

6. RELATIONS BETWEEN THE DUST AND SO_2 CONTENT

The daily average of dust and SO_2 content during the heating period in Wahnsdorf show very close relations. As can be seen from Figure 2, there was a quasi-parallel course of the dust and SO_2 content in the months of October to February 1965/6 and 1966/7.

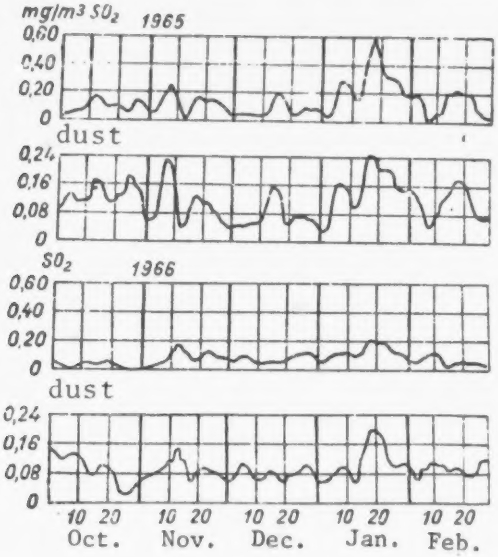

Figure 2. Dust and SO_2 concentration in Wahnsdorf, October - February, 1965/6, 1966/7 (smoothed values).

In order to compensate for accidental fluctuations in the graphic presentation, the measurements were smoothed by a 5-day running average. For the period from October to February, the correlation coefficients of the unsmoothed values are 0.85 for 1965/6 and 0.80 for 1966/7. In order to

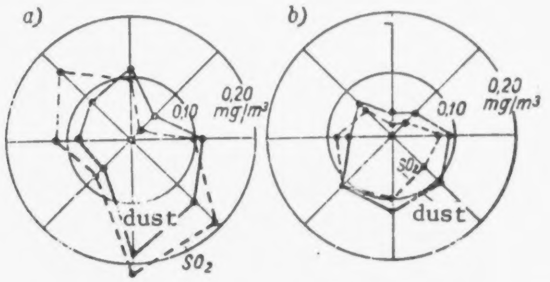

Figure 3. Dependence of the dust and SO_2 content on wind direction.
a. October 1965 - February 1966.
b. October 1966 - February 1967.

eliminate the conservation tendency of the meteorological diffusion processes, only the values of the first, third, fifth, etc. measuring days were considered for the correlation calculation. Also, as Figures 3 and 4 show, both trace materials show a dependence on wind direction

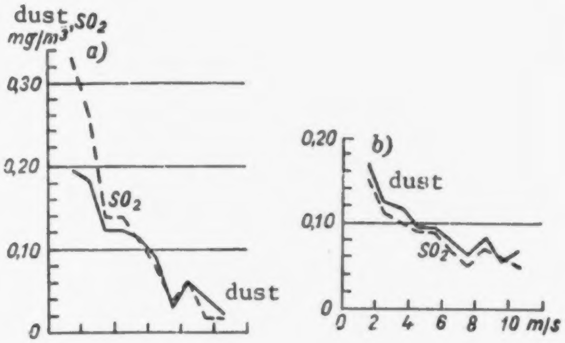

Figure 4. Dependence of the dust and SO_2 content on wind speed.
a. October 1965 - February 1966
b. October 1966 - February 1967

and wind speed. But it is worth noting that there are distinct differences between the two years in the concentration curve for the trace materials. These can be ascribed to differences in the diffusion processes. In the summer months there is no relation between the dust and SO_2 content. For the period of March to September, 1966, the correlation factor was 0.29. In the summer, the suspended dust consists not only of emissions from industry and house fires, but also in part of mineral dust which has been blown up. By convection, the dust enters into the higher layers of the air, from which it is only slowly removed by sedimentation. Sulfur dioxide is likewise transported by convection into higher air layers, but it is gradually converted to a sulfate aerosol, so that the SO_2 concentration is lower in the summer than in the winter.

7. CONCLUSIONS

Air pollution problems will increase in importance in all countries in the coming years as a result of the construction of new industrial centers and cities. Thus it should be the primary goal of all institutions interested in maintenance of air purity to aim at close technical-scientific and organizational cooperation in the planning and construction of air-chemical measuring networks, so as to guarantee that the measurements will be comparable.

ON THE CREATION OF A NETWORK OF POINTS FOR OBSERVATIONS OF ATMOSPHERIC POLLUTION IN THE GERMAN DEMOCRATIC REPUBLIC*

(ABSTRACT)

W. Warmbt (German Democratic Republic)

By decision of the Council of Ministers of the German Democratic Republic, the Hydrometeorological Service of the country has begun organizing studies of atmospheric pollution. This involves the participation of 13 meteorological stations located in different climatic zones outside the industrial and densely populated points at which measurements of ozone concentration and radioactivity had been made earlier.

The dust content is determined by filtering a known volume of air (approximately 60 m^3 per day) through membrane filters of Czechoslovak manufacture and weighing them before and after the filtering. The sulfur dioxide concentration of air was determined by the West-Gaeke method. The samples were collected 3 times a day for 1 hour, and in heavily polluted regions, for 30 minutes at an air suction rate of 50 l/hr.

The paper presents results of measurements of dust and sulfur dioxide concentrations carried out in Wahnsdorf in the winter periods (from October through February) of 1965-67. An almost parallel course in the variations of sulfur dioxide and dust concentrations with time was observed, with correlation factors of 0.80 and 0.85 between the quantities measured.

To study the air pollution in cities and industrial centers and in order to substantiate the maximum permissible norms for concentrations of noxious pollutants, specialized vans have begun to be used. Such mobile laboratories are provided with equipment for determining the concentration of sulfur dioxide,

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* This is a translation of the original Russian abstract accompanying this German paper.

carbon monoxide, carbon dioxide, hydrocarbons, nitrogen oxides, acrolein, and lead dust. The content of carbon monoxide, carbon dioxide, and hydrocarbons is recorded automatically. At the same time, the air temperature and humidity, visibility, and wind direction and velocity are measured.

CONTENT OF PHOTOOXIDANTS IN URBAN AIR

Yu. G. Fel'dman (USSR)

From Glavnoe Upravlenie Gidrometeorologicheskoy Sluzhby Pri Sovete Ministrov SSSR. (Chief Administration of the Hydrometeorological Service Under the Council of Ministers of the USSR.) "Meteorologisheskie Aspekty Zagryazneniya Atmosfery". (Meteorological Aspects of Air Pollution.) Sbornik dokladov na mezhdunarodnom simpoziume v Leningrade - Iyul' 1968 g. (Reports delivered at the International Symposium in Leningrad - July 1968.) Pod redaktsiey d-ra fiz.-mat. nauk M. E. Berlyanda. (Edited by Prof. M. E. Berlyand.) Gidrometeorolgicheskoe izdatel'stvo, Leningrad, p. 337-344, (1971). (Hydrometeorological Publishing House, Leningrad, (1971).)

Photooxidants are toxic products formed in atmospheric air from automotive and industrial emissions as a result of complex photochemical reactions stimulated by ultraviolet radiation of the sun. Products that, in addition to their biological activity, have a harmful effect on vegetation and reduce the visibility include ozone, peroxyacetyl nitrate (PAN), nitrogen oxides, formaldehyde, acrolein, free radicals, organic peroxides, finely divided aerosols, etc.

The pollution of air with oxidants, first noted at the end of the 1940's in Los Angeles (U.S.A.), has now become a serious problem for many American cities -- New York, Washington, San Francisco, etc.

In the U.S.A., scientific research institutions are looking intensively for the various chemical and physical agents responsible for the formation of photochemical smog. An extensive network of observation stations recording the level of urban air pollution has been created. In particular, in the state of California, there are 16 such stations, according to the data of which the maximum hourly concentrations of oxidants during the summer period of 1954 in Los Angeles were 0.63 mg/m^3, and the maximum 5-minute concentrations, 0.71 mg/m^3 (based on hydrogen peroxide). In 1965, the indices of air pollution by oxidants were approximately at the same level in that area.

The conditions favoring the formation of photochemical smog by reactive organic compounds and nitrogen oxides in the presence of a high air pollution level are an abundance of solar radiation, temperature inversions, and low wind speeds.

The oxidant concentrations are subject to great variations, but they follow certain patterns: as a rule, low nighttime concentrations are followed by a substantial increase in the early morning, then a peak takes place that remains over the entire course of solar radiance and disappears with the sunset. The highest concentrations are usually observed at noon.

Reports have been published on the oxidant pollution of the air reservoir of Italian cities. Kanitz [28] observed typical products of Los Angeles smog --

-- PAN and other homologs of this series, peroxypropionyl nitrate (PPN) and peroxybutyryl nitrate (PBN) in the air of Genoa.

Observations made by V. A. Popov [8] in Moscow, Baku, and Batumi demonstrate the presence of oxidants in the air of these industrial centers of the country. Thus, in Moscow, the maximum total concentration of oxidants based on Popov [8] in Moscow, Baku, and Batumi demonstrate the presence of oxidants in the air of these industrial centers of the country. Thus, in Moscow, the maximum total concentration of oxidants based on Popov's data was 0.1 mg/m^3, in Baku, 0.15 mg/m^3, and in Batumi, 0.04 mg/m^3.

The problem of the biological effect of oxidants as a new environmental factor has attracted the attention of researchers ever since the population of Los Angeles first reported mass complaints of eye irritation.

The irritant effect of oxidants is attributed by researchers primarily to the action of formaldehyde, acrolein and PAN.

According to the data of [35], in which the irritant effect of individual oxidants was studied experimentally, among the most sensitive persons, eye irritation is observed from exposure to formaldehyde in a concentration of 0.12 mg/m^3.

Studies by American authors prove that the inhalation of air containing even moderate concentrations of oxidants, determined in the atmosphere during periods of photochemical smog, are dangerous to persons with disturbances of the respiratory system.

Motley et al. [33] report the results of a study of the influence of photochemical smog on the respiratory function of 66 volunteer subjects, 46 of whom suffered from pulmonary emphysema. The authors came to the conclusion that the inhalation in the course of 2-3 days of air containing a large amount of oxidants, 0.4-1.47 mg/m^3, causes a decrease in the volume of maximum pulmonary ventilation and a decrease of the vital capacity of the lungs, the changes being more pronounced in persons suffering from pulmonary emphysema.

In a study of the possible relationship between asthma attacks and air pollution, a correlation was established in [34] between the number of attacks of this illness and the content of oxidizing substances in air.

A survey of the population of Los Angeles revealed that 40% of the persons questioned were worried by air pollution. Three-fourths of this number complained of eye irritation. One hundred thousand inhabitants of the state of California stated that air pollution affects their breathing. Some persons changed their residence because of air pollution [21].

The toxic effect caused by photochemical smog is thought to be substantially related to the action of ozone, which makes up an appreciable part of the total amount of oxidants.

It is well known that in low concentrations, ozone is observed almost everywhere in the surface layer of air as a natural factor. According to observations of ozone [41, 4, 5, 11 etc.], its concentrations in the layer up to 2 km from the earth's surface are highly variable and depend on the time of year, latitude, time of day, and local conditions. These concentrations range from thousandths of a milligram to 0.15 mg/m^3 and amount to an average of 0.01-0.04 mg/m^3.

The irritant effect of ozone in concentrations of 0.2-1.0 mg/m^3 on the mucosa of the eyes is noted in [3, 7, 29, 34].

According to the data of [22, 13, 38], the ozone concentration in the air of Los Angeles climbed to 0.6-1.0 mg/m^3 during a photochemical smog.

According to many researchers, the inhalation of air containing increased ozone concentrations causes a decrease in the resistance of the organism. Thus, after a 3 hour exposure of white mice to ozone in concentrations of 0.1-0.98 mg/m^3, followed by infection with streptococcus aerosol, a 25% increase in death rate among the animals was observed as compared with the control [19].

A similar effect of increased death rate of animals is noted in [38], based on data of exposure of newborn mice to ozone in concentrations of 0.19-0.38 mg/m^3 in 7-hour periods in the course of three weeks.

The action of PAN and its homologs on man was studied only in brief exposures. These compounds affect the respiratory function in the manner of other oxidants. In essentially healthy students who inhaled PAN in a concentration of 1.5 mg/m^3 for 5 min, a statistically significant increase in oxygen consumption was observed. In a concentration of 2.5 mg/m^3, this substance has a more irritant effect on eyes in a 12-min exposure. However, the role of PAN among other lacrimators is not completely clear, since it is less toxic in pure form than in the composition of photochemical smog [36].

The object of the present study was a further accumulation of data on the total content of oxidants and individual oxidizers (ozone and nitrogen dioxide) in the air of a large city. Another objective was to study the vertical scattering of oxidants in order to obtain a comparative picture of the pollution of atmospheric air at the level of the lower and upper floors of high-rise apartment buildings, which are being widely incorporated into typical residential construction.

Literature data on the content of atmospheric pollutants at different levels from the ground are scarce. Attention is drawn to the studies made by M. K. Kharakhinov [10], which showed that the carbon monoxide concentrations at the height of the 22nd floor of a high-rise building in Moscow are almost the same as the concentrations at a level of 1 m from the ground.

Analogous data of vertical distribution of dust were obtained on the same site by G. I. Sidorenko [9]. He also found that at the height of the 22nd floor, the number of microorganisms is 63.6% of the number at a height of 1.5 m above ground.

According to [2], in a study of the content of noxious pollutants in the air above a motor roadway and in residential apartments on the first three floors, the highest level of carbon monoxide concentrations was associated with the upper floors. The concentrations of nitrogen oxides in the street and in third-floor apartments were the same.

To determine the total amount of oxidants, we used a method based on the reaction of oxidation of phenolphthalin to phenolphthalein [31]. The reddish-violet color formed was compared on the spot with an artificial scale. The sensitivity of the method is 0.1 μg per 5 ml of the solution studied.

The ozone content was determined spectrophotometrically with the reagent dihydroacridine [6]. The method is based on measurement of the optical density of an ethanol solution of acridine, formed by reacting ozone with dihydroacridine.

To determine the nitrogen dioxide, the air was passed through a solution of potassium iodide, then a standard analysis using Griess reagent was carried out on a photoelectrocolorimeter [1]. In addition to the determination of the indicated substances, the air was analyzed for the content of one of the most toxic components of automotive exhaust, carbon monoxide, by means of a TG-5 gas analyzer [1].

The subject of this study were two 12-story apartment buildings located next to roadways with no gap between the road and the safety* line. One of the buildings (object *a*) was situated next to a highway with a traffic flow of 2000-2400 cars per hour, and the other (object *b*), with 1500-1800 cars per hour. The number of cars was counted visually.

The air samples were collected during the summer-autumn period of 1967 directly on the highway and on balconies on the 8th and 12th floors.

The collection of air samples was associated with the recording of meteorological conditions, intensity of solar radiation, and rate of traffic flow. The period of the studies was associated with sunny weather and occasional variable cloudiness without precipitation; the air temperature was 12.8-26°, the relative humidity 27-73%, the wind velocity 0.2-2.1 m/sec, and the pressure 738-765 mm. The intensity of total solar radiation ranged from 0.02 to 1.08 cal/cm^2 min. On the objects studied, 245 determinations of the total amount of of oxidants, 66 of ozone, 135 of nitrogen dioxide and 287 of carbon monoxide were carried out (total of 733) tests).

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* Translator's note: Literally the red line.

Fig. 1 gives average and maximum values of the concentrations of these ingredients, determined in the street and on balconies of the 8th and 12th floors. As is evident from the figure, the highest oxidant concentrations were observed on object *a*. Although these concentrations decrease somewhat with height, the indices obtained on all the levels have similar values (0.166, 0.134, and 0.125 mg/m^3 respectively).

The maximum oxidant concentrations were found during the period of 11 A.M.-1 P.M., when the highest intensity of solar radiation was observed (1.08 cal/cm^2 min). As the intensity of solar radiation diminishes, the oxidant concentrations decrease, indicating the photochemical nature of their origin.

In cloudy weather and particularly during rains, the oxidant content of air decreased to 0.01-0.02 mg/m^3.

The nitrogen dioxide concentrations on object *a* remained unchanged up to the level of the 12th floor, exceeding the MPC for this residential area. As far as the indices of ozone content are concerned, their hygienic significance is difficult to evaluate because of a lack of a maximum permissible concentration for atmospheric ozone. However, we note that the ozone concentrations in the street as well as at the height of the 12th floor (0.03-0.035 mg/m^3) are twice as large as its odor threshold, 0.015 mg/m^3, according to M. E. Eglite [11].

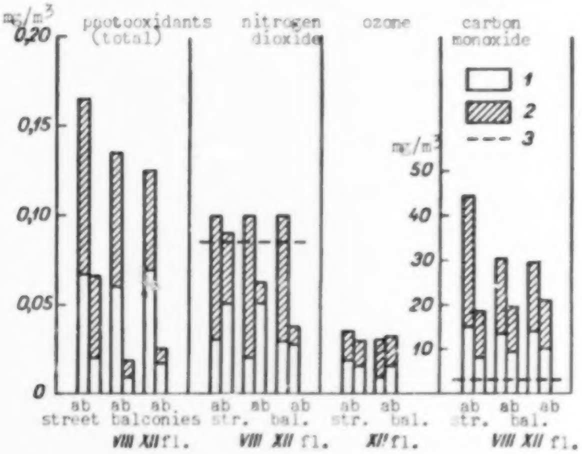

Fig. 1. Concentration (mg/m^3) of photooxidants and carbon monoxide in the air of motor roadways for objects a and b.

1 - average concentrations, 2 - maximum concentrations, 3 - MPC (maximum one-time value for atmospheric air).

Thus on the major motor roadways surveyed in this cycle of observations, the pollution of air with oxidants and nitrogen dioxide extends to the higher floors of high-rise residential buildings.

Familiarization with the literature on this problem shows that the limited number of substances forming a photochemical smog were identified with a sufficient degree of accuracy.

Experimental studies should be made on the threshold and inactive concentrations of oxidants by using the sensitive tests employed for standardizing atmospheric pollutants in the USSR. It is also very important to determine whether oxidants in the concentrations recorded in the air of industrial centers of the country are biologically active.

In this connection, future development of research should be conducted in the following principal directions:

(a) study of the photochemical pollution of urban air in different climatic zones, taking the local meteorological conditions into account;

(b) simulation of the concentrations of the main ingredients of photochemical smog observed in nature, for the purpose of studying the effect on the organism of animals and man;

(c) development of ameliorative hygienic, sanitary-planning, sanitary-technical and other measures aimed at protecting the air of cities.

A particularly timely step toward a successful solution of the formulated problem is the participation of agencies of the Meteorological Service in full-scale observations in different cities.

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Ju. G. FELDMAN

ON PHOTOOXIDANT CONTENT IN THE TOWN AIR*

Photooxidants are toxic products which are formed in atmospheric air from motor transport and industry emissions as a result of complex chemical reactions stimulated by ultraviolet solar radiation. To these products possessing the ability to harm vegetation and to decrease visibility along with biological activities refer: ozone, nitrogen oxides, peroxyacetylnitrate (PAN), organic peroxides, formaldehyde, acrolein, fine-dispersed aerosols, free radicals, etc.

The object of this work was the accumulation of data on summary content of oxidants, and of ozone and nitrogen dioxide separately, in the air of the city. Besides, the determination of concentrations of one of the most toxic components of automobile exhaust, i. e. carbon oxide, was carried out. The sum of oxidants was determined on reaction with phenolphthalin, ozone with dihydroacridine, nitrogen dioxide with Griess reagent, and carbon oxide on gas analyzers TG-5, respectively.

The content of these substances in the air was studied both directly on roads with motion intensity of 1500—2400 cars an hour, and on balconies of the 8th and 12th storeys of the houses situated on these roads.

The obtained results show that oxidant concentrations on all the levels are close quantities (0.166, 0.134, and 0.125 mg/m^3, respectively), though they decrease with height to some extent.

Maximum oxidant concentrations were found in the period of the most intensive solar radiation, that is from 11 a. m. till 1 p. m. Oxidant content in the air was decreased to 0.01—0.02 mg/m^3 at dull weather and during the rain, especially. Nitrogen dioxide concentrations remained unchangeable up to the level of the 12th storey surpassing maximum permissible concentration for residential area. Ozone concentrations amounted to 0.03—0.035 mg/m^3 both at the bottom and at the height of the 12th storey which exceeds its smell threshold of 0.015 mg/m^3 by factor of two. In this connection, limited and non-acting photooxidant concentrations have to be determined experimentally on tests usually used for pollution normalization.

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* Editor's note: The abstract is presented as given in English with the original Russian article but has been slightly edited.

STUDY OF AIR POLLUTION AND ATMOSPHERIC PRECIPITATION RESULTING FROM ARTIFICIAL MODIFICATION OF CLOUDS

Sh. G. Gavasheli (USSR)

From Glavnoe Upravlenie Gidrometeorologicheskoy Sluzhby Pri Sovete Ministrov SSSR. (Chief Administration of the Hydrometeorological Service Under the Council of Ministers of the USSR.) "Meteorologisheskie Aspekty Zagryazneniya Atmosfery". (Meteorological Aspects of Air Pollution.) Sbornik dokladov na mezhdunarodnom simpoziume v Leningrade - Iyul' 1968 g. (Reports delivered at the International Symposium in Leningrad - July 1968.) Pod redaktsiey d-ra fiz.-mat. nauk M. E. Berlyanda. (Edited by Prof. M. E. Berlyand.) Gidrometeorolgicheskoe izdatel'stvo, Leningrad, p. 345-348, (1971). (Hydrometeorological Publishing House, Leningrad, (1971).)

At the present time, extensive work is being done on the prevention of damage done by hail, and on the development of precipitation and the scattering of fogs by means of various chemical agents. The development of methods of modification of meteorological phenomena and the improvement of existing ones constitute one of the major problems of modern science.

In recent years, extensive work on protection of vineyards from hail damage has been conducted in Georgia and other republics. The chief reagent used for hail prevention at the present time is lead iodide. Lead is widely distributed in nature: in air, soil, water, plants, and the human and animal organisms.

The lead content of the atmosphere and atmospheric precipitation has been studied by V. I. Baranov, I. I. Vilenskiy, M. N. Bykov, A. S. Zykova, V. A. Ryazanov, O. P. Shalamberidze, and others. The effect of lead on public health and its action on the organism have been studied by O. S. Yesyutina, A. A. Letavet, Ya. Z. Matusevich, N. I. Tarasenko, M. K. Khachatryan, and L. N. Khvil'nitskaya.

It is well known that the chief sources of air pollution with lead are ferrous metallurgical plants, lead-ore dressing, printing type, and storage battery plants, etc.

Our aim was to study the degree of pollution of the atmosphere resulting from cloud modification. To this end, sampling of atmospheric precipitation and air and their subsequent analysis were organized.

The chief requirement for a sampling instrument is that the latter itself must not introduce any additional contaminants into the samples being collected. We therefore constructed a special "precipitation collector" instrument made of 8-10 mm plexiglas.

The precipitation collector consists of two main parts, a receiving vessel and a lid. The receiving vessel is in the shape of a quadrangle with a receiving surface of 58 x 60 cm. The lid of the precipitation collector is a rectangle whose lower side has a half-thickness groove for tightly covering the receiving vessel.

Before the precipitation, the lid is removed by means of a special handle, and placed at an angle in the rear portion of the receiving vessel so that the atmospheric precipitation falling on the inner side of the lid flows directly into it. Thus, the lid performs a dual function: first, it protects the receiving part of the vessel from dust in rainless weather, and secondly, during precipitation, it almost doubles the receiving surface of the instrument, permitting the collection of a large number of samples of atmospheric precipitation.

The collected samples are poured from the receiving vessel through a drainage hole into a special polyethylene vessel, which is sent to a chemical laboratory for analysis.

During the expeditions, samples of air were taken for chemical analysis and determination of their lead content. The method consisted in the following: air was pumped through a Kipp instrument, containing doubly distilled water, with the aid of the so-called dust accumulator of type UA-T-80, which directly measured the total amount of drawn air. The lead particles present in the air remained in the doubly distilled water on passing through it.

There are a good number of methods of determination of lead in different samples. A given method is used as a function of the problem at hand. For our purposes, i.e., in determining lead in atmospheric precipitation and in air, the sulfide method was employed.

The protection of vineyards from hail damage on the territory of Georgia is carried out mainly in Kakhetya in the valley of Alazini River, this being the region with the most advanced viticulture in the Republic.

The installations for introducing the reagents into the clouds were mainly located on the mountain tops and slopes of the Caucasian and Gomborskiy ridges. The bulk of the reagents introduced into the clouds precipitate out in the central part of the Alazini valley. For this reason, most of the precipitation collectors were placed in this region.

In 1964-67, 210 samples of atmospheric precipitation were collected in which the lead content was determined. From the data obtained it follows that the highest average lead concentration in atmospheric precipitation was observed in Chotori in 1966 and amounted to 0.0054 mg/l, whereas in Telavi it was 0.0014 mg/l, and in Leliane, 0.0023 mg/l.

The maximum amount of lead in single samples of atmospheric precipitation was also obtained in Chotori on 12 June 1968 and amounted to 0.0200 mg/l. In

Telavi, the maximum amount was observed in 1967 and amounted to 0.0035 mg/l (in Chotori, no atmospheric precipitation was collected in 1967).

On 12 June 1966 in Kakhetya there was a total cloud cover, a thunder storm began at approximately 11:30 A.M. and continued until 3-4 P.M. There was a pouring rain, and the wind was gentle in the northern and northwestern directions. On that day, cloud modification work was being carried out by expeditions of the Geophysical Institute of the Academy of Sciences of the Georgian SSR and the Transcaucasian Scientific Research Hydrometeorological Institute.

Similar cases of increase of lead content in precipitation as a result of cloud modification were obtained on other days, for example, 25 May and 24 June 1966.

This direct relationship between cloud modification and increase of lead concentration in precipitation was not always observed. It is probable that various factors are involved here: the amount of lead introduced into the clouds, meteorological conditions, number of precipitation collectors located on the territory studied (for observation of the region of maximum pollution), etc.

It is of interest to note that an increased lead concentration (0.0214 mg/m^3) was also obtained on 13 and 14 June 1966 in atmospheric air. Cloud modification was carried out on 12 and 13 June.

It is necessary to compare the data obtained with the maximum permissible concentration adopted in the USSR.

The maximum permissible concentrations (MPC) of lead in atmospheric precipitation have not yet been established. It should also be noted that not enough studies have been made thus far on the determination of lead in atmospheric precipitation. However, one has to assume that the MPC of lead in atmospheric precipitation should be higher than in drinking water (0.1 mg/l). On the basis of the above, it may be concluded that despite the fact that in some samples the lead content of atmospheric precipitation is fairly high, it still remains below the maximum permissible concentrations.

The situation is different in the case of the amount of lead in atmospheric air. In most of the collected samples, the amount of lead in the atmospheric air of Kakhetiya was found to be greater than the MPC, and on some days, much greater. For example, on 13 June 1968 in Chotori, the value of 0.0214 mg/m^3 was obtained, whereas the MPC in the USSR for populated areas is 0.007 mg/m^3.

Thus, as a result of artificial modification of clouds, the amount of lead in atmospheric precipitation increases, but it still remains below the maximum permissible concentration. As far as the air is concerned, the amount of lead increases considerably, and at some individual points exceeds the PMC by a factor of 30-35.

Sh. G. GAVASHELI

INVESTIGATION OF AIR POLLUTION AND ATMOSPHERIC PRECIPITATION AS A RESULT OF ARTIFICIAL MODIFICATION OF CLOUDS*

In the last few years work on modification of meteorological phenomena is performed on a large scale. On considerable areas of Georgia protection of agricultural crops from hail is fulfilled. Lead iodide is used as a main crystallizing reagent employed in artificial modification of clouds. It is interesting to evaluate to what extent air and precipitation are polluted after introduction of this reagent into clouds.

From 1964 Scientific-research Hydrometeorological Institute of the Caucasus carries special investigations with purpose of determination of air pollution level at modification of clouds. Air and precipitation sampling and their analysis on lead content is done. For precipitation sampling a special sampler was constructed which consists of collector with a lid. The latter protects the collector from the dust and during the precipitation increases the sampling area nearly twice. This allows to collect large amounts of precipitation for the analysis on lead content. For air sampling Kipp's instrument is used through which investigated air is passed.

Air and precipitation samples were taken during expeditions in Georgia while conducting artificial modification of clouds. On the whole, 210 samples of precipitation and 20 air samples were analyzed for 1964—67. Obtained data analysis shows that lead amount in precipitation increases at modification, but still remains less than maximum permissible concentration. The largest mean lead concentration in precipitation was observed in Chotory on the 12th of June, 1966. It amounts to 0.02 mg/l. As to the air, the lead amount in it considerably increases at modifications, sometimes surpassing maximum permissible concentration in 30—35 times. On June 13, 1966 in Chotory the measured lead concentration in the air was equal to 0.024 mg/m^3 and exceeded maximum permissible concentration in about 34 times.

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* Editor's note: The abstract is presented as given in English with the original Russian article.

MICROCLIMATIC CHARACTERISTICS AND HYGIENIC EVALUATION OF THE RELATIVE EMPLACEMENT OF INDUSTRIAL AND RESIDENTIAL COMPLEXES IN THE REGIONS OF SIBERIA

L. I. Koldomasov and M. T. Zenin (USSR)

From Glavnoe Upravlenie Gidrometeorologicheskoy Sluzhby Pri Sovete Ministrov SSSR. (Chief Administration of the Hydrometeorological Service Under the Council of Ministers of the USSR.) "Meteorologisheskie Aspekty Zagryazneniya Atmosfery". (Meteorological Aspects of Air Pollution.) Sbornik dokladov na mezhdunarodnom simpoziume v Leningrade - Iyul' 1968 g. (Reports delivered at the International Symposium in Leningrad - July 1968.) Pod redaktsiey d-ra fiz.-mat. nauk M. E. Berlyanda. (Edited by Prof. M. E. Berlyand.) Gidrometeorolgicheskoe izdatel'stvo, Leningrad, p. 349-351, (1971). (Hydrometeorological Publishing House, Leningrad, (1971).)

A very important factor in the modernization, design, and construction of cities and in the planning of new industrial centers is a scientifically valid consideration of microclimatic characteristics of the region. Most important is the influence of the topography, which is usually responsible for the generation of local air circulation, and largely determines the conditions of transport of noxious emissions of industrial plants toward residential areas and hence the degree of atmospheric pollution. Under dissected relief conditions, the wind roses differ markedly from one another even at closely situated meteorological stations. For this reason, in solving urban construction problems, particularly in determining the relative location of industrial and residential complexes, it is important to study the local microclimatic characteristics. Their underestimation in many cities of the country has led to a considerable increase in air pollution.

The Applied Climatology Section of the Novosibirsk Branch of the Hydrometeorological Center (formerly, a branch of the Scientific Research Institute of Aeroclimatology) since 1958 has been conducting (in collaboration with the Novosibirsk Scientific Research Sanitation Institute) extensive microclimatic and sanitary-hygienic surveys of certain cities in which meteorological observations and the collection of air samples are combined at 12 to 15 stationary points. At these points, the concentrations of dust, soot, and a number of gaseous ingredients are regularly determined.

As we know, data given in climatological handbooks do not include a whole complex of meteorological data that are required for characterizing the conditions of propagation of noxious emissions. In particular, the handbooks do not contain information on the frequency of dangerous velocities and directions of the wind (average and for the observations periods), or on the continuous duration of periods with calms and with gentle winds.

The propagation and concentration of impurities are strongly affected by temperature inversions, which limit the ascent and dispersal of noxious emissions from plant stacks. In winter, inversions in Siberia are observed almost daily, their frequency in the cold half of the year amounts to 90% or more, and their thickness frequently exceeds 500-600 m. The distance between the points of radial sounding in Siberia is very large. In this connection, studies were made for the purpose of working out methods of calculation of the frequency of inversions that are dangerous from the standpoint of atmospheric pollution, using surface and pilot balloon observations. In particular, satisfactory relationships were observed between the average and maximum frequency of winter morning inversions and the frequency of days with a weak mean daily wind velocity.

The problems of relating such observations (1-2 years) on the wind velocity to long-term data of a local meteorological base station were studied.

In work over rugged terrain, a study was made of the distortion of air currents flowing from the plants to sites intended for residential construction, primarily in the presence of weak and moderate wind velocities. In addition, the effect of different types of industrial sites and of residential buildings on the air pollution was investigated.

To determine the influence of noxious emissions on the sanitary living conditions, a questionnaire was submitted to the population, and the state of health of the children living in various parts of the city was surveyed.

In one city with a developed metallurgy, a metallometric survey was made within a 30 km radius in collaboration with geologists, which showed that as a rule, increased metal concentrations in the soil are observed in zones where high average concentrations of the metal are present in the dust collected from the air.

Correlation and analysis of the data of the studies made it possible to supply a comprehensive microclimatic and hygienic evaluation of the territory of certain cities to the planning and design agencies. Variants of the relative emplacement of industrial facilities and populated areas were evaluated, boundaries of sanitary protective zones were refined, and recommendations were given on the selection of sites for public rest and recreation areas and for planting greenery.

L. I. KOLDOMASOV, M. T. ZENIN

MICROCLIMATIC PECULIARITIES AND HYGIENIC ESTIMATION OF MUTUAL LOCATION OF INDUSTRIAL AND RESIDENTIAL COMPLEXES IN THE REGIONS OF SIBERIA*

Scientifically valid calculation of local conditions and microclimatic features has a great significance for reconstruction, planning and building of existing towns, and projection of new industrial centres.

It is very important to take into account the influence of terrain relief, especially in complex conditions of mountain and foot-hill regions where local winds usually occur which determine transfer conditions and the level of air pollution by industrial plants.

As the result of integrated microclimatic and sanitary-hygienic inspection of some Siberian towns it has been found that underestimation of local features often leads to substantial pollution of residential regions by noxious industrial emissions which affects human health.

As investigations show the highest air pollution by nitrogen oxides, phenol, dust, sulphur oxide, and other ingredients is usually marked at calm or light (up to 2 m/sec) wind and in the presence of ground and elevated inversions with thickness not less than 300 m. If such weather continues more than two days the concentrations of noxious emissions above maximum permissible values are observed, as a rule, on the whole territory of the town.

For the estimation of air pollution level and discharge distributions on town territory and for the definition of boundaries of sanitary-protecting zones for main enterprises, it is necessary to carry out integrated microclimatic and sanitary-hygienic inspection during at least two cycles.

In towns with developed metallurgical industry, copper, nickel, iron, manganese, zinc, etc., are accumulated in the soil during many years. The distribution of separate ingredients content in the soil on the territory of the town and its suburbs can be revealed with the help of metallometric survey and in such a way it is possible to receive indirect characteristics of air pollution.

Microclimatic investigations are carried out in new industrial regions on the basis of which recommendations are given on the choice of the best variants of mutual location of industrial and residential complexes.

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* Editor's note: The abstract is presented as given in English with the original Russian article, but has been slightly edited.

NUMERICAL CHARACTERISTICS OF METEOROLOGICAL CONDITIONS ASSOCIATED WITH PERIODS OF HEAVY ATMOSPHERIC POLLUTION IN WESTERN SIBERIA

I. A. Shevchuk and L. I. Vvedenskaya (USSR)

From Glavnoe Upravlenie Gidrometeorologicheskoy Sluzhby Pri Sovete Ministrov SSSR. (Chief Administration of the Hydrometeorological Service Under the Council of Ministers of the USSR.) "Meteorologisheskie Aspekty Zagryazneniya Atmosfery". (Meteorological Aspects of Air Pollution.) Sbornik dokladov na mezhdunarodnom simpoziume v Leningrade - Iyul' 1968 g. (Reports delivered at the International Symposium in Leningrad - July 1968.) Pod redaktsiey d-ra fiz.-mat. nauk M. E. Berlyanda. (Edited by Prof. M. E. Berlyand.) Gidrometeorolgicheskoe izdatel'stvo, Leningrad, p. 352-356, (1971). (Hydrometeorological Publishing House, Leningrad, (1971).)

Studies of the meteorological conditions associated with periods of heavy atmospheric pollution in industrial cities of Siberia are being conducted at the Novosibirsk Regional Hydrometeorological Center for the purpose of working out recommendations of methods to be used for warning interested agencies of impending periods of possible heavy pollution.

The material used in this study consisted of regular daily observations of the propagation of emissions from industrial plants and from transport in several cities of Siberia. Data for 1962-64 and 1966-67 were used.

The observations were made at stationary points of the cities and along special routes on trucks. Samples of air for analysis of dust, soot, nitrogen oxides, carbon monoxide, and sulfur dioxide were taken at the earth's surface and high in the air by using a helicopter. The data obtained were used to determine the average and maximum concentrations of the ingredients at various points of the cities for every 24-hour period.

The period of heavy atmospheric pollution was considered to include cases in which in a specific city, the highest average concentrations of two or more ingredients were simultaneously retained for over two consecutive days. Thirty-three such periods were isolated during the periods considered, 15 during the warm time of the year and 18 during the cold. Overall, periods of heavy pollution amounted to 208 days.

Based on data of meteorological and aerological observations, the average and maximum wind velocities, vertical thermal gradient of the layer and average horizontal pressure gradient on the ground and at a level of 850 mb, calculated as a Laplacian, were considered in the lower 2-kilometer layer for each day of the isolated period (the average wind velocity taken in the layer was the arithmetic average of velocities at 6 levels -- ground, 100, 200, 500, 900 m above the earth's surface and 1500, 2000 above sea level).

Analysis of the material showed that in the regions of Siberia, a prolonged and appreciable increase of the ingredient concentrations near the earth's surface is most likely to occur under conditions of a low-gradient baric field in the presence of a stably stratified air mass; this generally confirms the results of studies on other territories.

The baric field in the region of Siberian cities for cases involving an increase in atmospheric pollution was diverse in character (anticyclonic, cyclonic and hyperbolic points). However, it almost always caused an attenuated air transport to a height of 1.5-3.0 km.

In the plains of Western Siberia, for cities with plant stack heights from 20 to 100 m, the specific characteristics of the state of the lower atmospheric layer during periods of heavy air pollution had the following values.

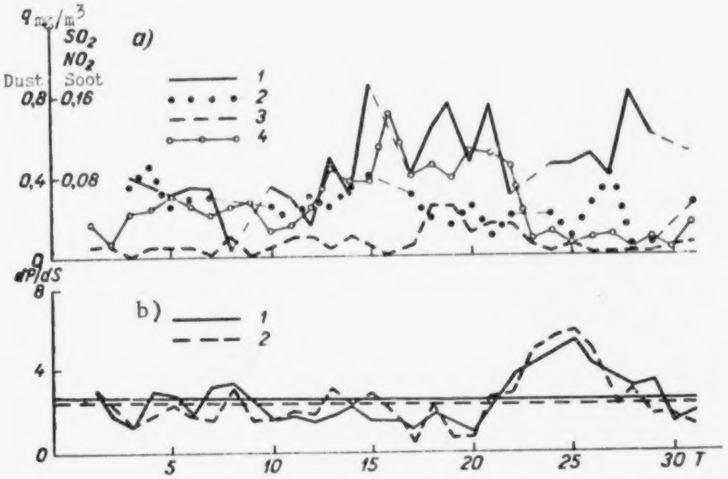

Fig. 1. Variation of average concentrations of ingredients (a) and average baric gradients (b) for February 1967.
(a) concentrations: 1 - nitrogen oxides, 2 - sulfur dioxide, 3 - soot, 4 - dust;
(b) baric gradients: 1 - on the earth's surface, 2 - on 850 mb. surface

1. The average horizontal baric gradient at sea level was 0.75-2.5 mb/500 km, and on the 850 mb surface, 0.75-3.0 mb/500 km. Simultaneous analysis of average concentrations and average horizontal baric gradients in February and July (Fig. 1, 2) clearly shows a relationship between high concentrations and minimum gradients.

2. The average wind velocity in the layer up to 2 km is preserved in the range from 2 to 6 m/sec. The maximum velocity at one of the levels in

the layer is not more than 7 m/sec in summer and not more than 10 m/sec in winter.

3. A stable stratification in the 2-kilometer layer is preserved during an entire 24-hour period. At night, a surface inversion is bound to be observed. In winter, the thickness of the inversion layer is not less than 800 m, and the maximum temperature gradient in it is not less than 2°C./100 m. In summer, the thickness of the inversion layer is not less than 100 m, and the maximum temperature gradient in it is not less than 1°C./100 m.

Differences in the meteorological characteristics of the 2-kilometer layer of the atmosphere during periods of heavy pollution in winter and summer are due to the temperature difference between the emissions and the ambient air in different seasons of the year.

When the above conditions are preserved, high concentrations of gaseous ingredients are observed during the entire period. The maximum dust concentration at the earth's surface is observed when, after a period of stagnation, the wind begins to intensify at the earth's surface, and the inversion, breaking up at the bottom, changes into an elevated inversion. When a frontal weatherparting passes through a city, even under conditions of a diffuse baric field, a decrease in the concentrations is observed both on the ground and high in the air, independently of whether there is or is not any precipitation as the front passes.

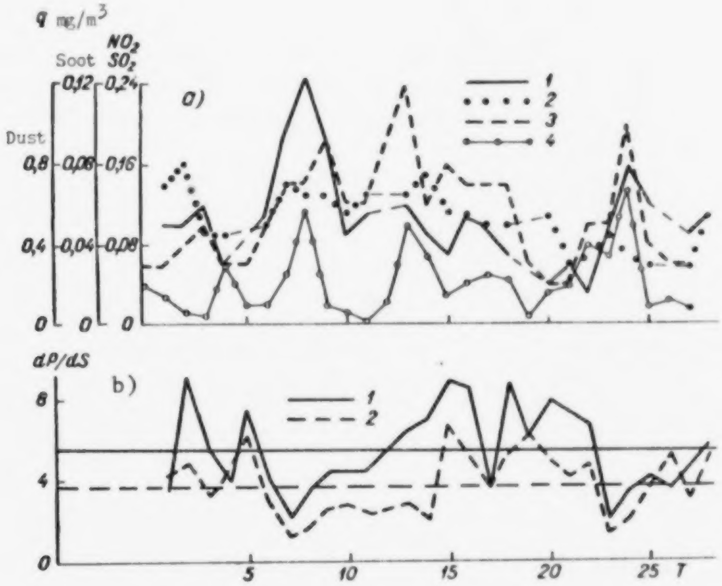

Fig. 2. Variation of average concentrations of ingredients (a) and average baric gradients (b) for July 1967.
Notation same as in Fig. 1.

To determine the probability of appearance of conditions promoting an increase in atmospheric pollution, results of daily radiosonde observations for 5 years (1962 through 1966) for Novosibirsk were processed. Graphs of the probability of appearance of unfavorable stratification under atmospheric stagnation conditions were plotted for different seasons of the year (Fig. 3).

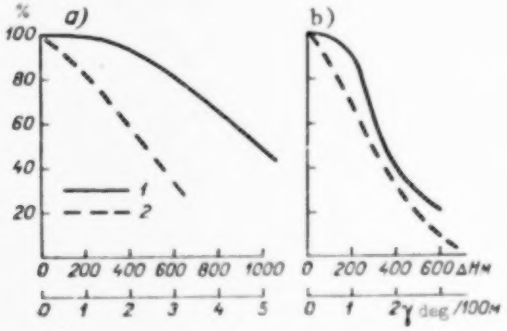

Fig. 3. Probability of appearance of unfavorable stratification under atmospheric stagnation conditions in winter (a) and summer (b).

1 - thickness of inversion layers, 2 - temperature lapse rate in inversion layers.

The numerical characteristics of horizontal baric gradients, wind velocity and intensity of inversion layers during periods of heavy atmospheric pollution, shown in Table 1, may be used for forecasting conditions causing a dangerous increase of the concentrations. The basis of such forecasting is the calculation of a future baric field on the ground and at the 850 mb level, and also the analysis of the variation in the stratification of the air mass.

Table 1

Meteorological Characteristics of the State of the Atmosphere Promoting a Considerable Increase in the Concentration of Industrial Emissions near the Earth's Surface.

Season	Laplacian		Wind Velocity in in 2 km layer, m/sec		Maximum Temperature Lapse Rate in Layer, deg/100m	Thickness of Stable Layer, m
	Ground	AT_{850}	Average	Maximum		
In Summer	0,75—2,5	0,75—3,0	2—6	≤7	≥ −1	>100
In Winter				<10	≥ −2	>800

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I. A. SHEVCHUK, L. I. VVEDENSKAYA

NUMERICAL CHARACTERISTICS OF METEOROLOGICAL CONDITIONS WHICH ACCOMPANY THE PERIODS OF STRONG AIR POLLUTION IN WESTERN SIBERIA*

On the basis of regular observations of pollutant distribution on the territories of Siberian towns in 1962—64 and 1966—67, the periods are marked out when the sharp increase of background concentrations of pollutants was observed. The analysis of meteorological and aerological observations during these periods showed that the background concentration increase is connected with stagnant atmospheric state. In the article there are stated particular numerical characteristics of meteorological elements near the ground surface, baric gradients, mean and maximum wind velocities, and stratification of the lower layer in the periods of strong air pollution.

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*Editor's note: The abstract is presented as given in English with the original Russian article.

EXPERIENCE IN SIMULATING THE PROPAGATION OF NOXIOUS SUBSTANCES IN THE SURFACE ATMOSPHERIC LAYER OVER PLANT SITES AND SURROUNDING GROUNDS

V. M. El'terman (USSR)

From Glavnoe Upravlenie Gidrometeorologicheskoy Sluzhby Pri Sovete Ministrov SSSR. (Chief Administration of the Hydrometeorological Service Under the Council of Ministers of the USSR.) "Meteorologisheskie Aspekty Zagryazneniya Atmosfery". (Meteorological Aspects of Air Pollution.) Sbornik dokladov na mezhdunarodnom simpoziume v Leningrade - Iyul' 1968 g. (Reports delivered at the International Symposium in Leningrad - July 1968.) Pod redaktsiey d-ra fiz.-mat. nauk M. E. Berlyanda. (Edited by Prof. M. E. Berlyand.) Gidrometeorolgicheskoe izdatel'stvo, Leningrad, p. 357-361 (1971). (Hydrometeorological Publishing House, Leningrad, (1971).)

Interesting results on the simulation of air currents in the atmosphere have been obtained at the A. I. Voyeykov Main Geophysical Observatory and Moscow University [2].

Work on simulation of the processes occurring in the surface atmospheric layer is also being conducted at the All-Union Central Scientific Research Institute of Labor Protection, All-Union Central Trade Union Council [7]. Since 1963, the concentrations of noxious substances in the atmosphere have been determined here in a wind tunnel on models of newly designed industrial plants and of those undergoing modernization. Studies have been made on two major chemical works, plastics factories and aluminum plants, two synthetic rubber plants being modernized, a blocked building of large width, and other plants.

The studies performed made it possible to find better solutions to the design of plants, and to determine the sites, heights and maximum amount of emissions of polluted air for which the purity of the surface atmospheric layer required by sanitary norms is maintained.

A simulation procedure based on the following assumptions was used in this study. From the equation of turbulent diffusion describing the process of impurity transport in atmospheric air, the following similarity criterion may be proposed:*

$$\frac{vL}{K} = \text{idem}, \qquad (1)$$

where v is the flow velocity, L is the characteristic dimension, and K is the coefficient of turbulent exchange.

According to [6], the following relation applies to the coefficient of turbulent exchange K.

$$K \sim \varepsilon^{1/3} l^{4/3}, \qquad (2)$$

where ε is the dissipation energy.

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* As was shown by the experiments, relative to the Reynolds criterion, the process studied is self-similar for Re > 2000.

From (2) and (1), assuming that $\frac{L}{l}$ = constant, we find

$$\frac{vL}{\varepsilon^{1/3}l^{4/3}} = \frac{v}{\varepsilon^{1/3}l^{1/3}} = \text{idem}. \tag{3}$$

Hence, the scales of velocities, lengths and dissipated energy may be mutually related by the expression

$$c_v = (c_\varepsilon c_l)^{1/3}. \tag{4}$$

Here $c_v = \frac{v_m}{v_f}$, $c_\varepsilon = \frac{\varepsilon_m}{\varepsilon_f}$, $c_l = \frac{l_m}{l_f}$ are the scales of the velocities, magnitudes of dissipated energy, and lengths; subscripts m and f designate the model and full scale, respectively.

The same type of result may be obtained directly from the condition of equality of the Kármán criterion for the model and full scale [4]

$$\mathrm{Ka} = -\frac{v'}{v},$$

where v' are fluctuation velocities.

Since $v' \sim \varepsilon^{1/3} l^{1/3}$, we obtain the condition (3). On the basis of this function, it is necessary to calculate the model for the case of isothermal flow. From the two scales chosen with a view to creating the most favorable conditions for carrying out the experiments, Eq. (4) permits one to determine the third scale.

The amount of energy dissipated in the air flow may be changed by introducing kinetic energy into the volume from which air is withdrawn by the wind tunnel [7].

In the case of nonisothermal flow, when the temperature or density of the gases ejected from the stacks is not equal to the density of the ambient air, or when a nonuniform distribution of temperatures takes place in the ambient medium itself, the simulation should, in addition, satisfy the Archimedes criterion, which is the ratio of the bouyant forces to the forces of inertia.

$$\mathrm{Ar} = \frac{gl}{v^2}\frac{\Delta\gamma}{\gamma}. \tag{5}$$

If the difference in density is only a consequence of the temperature difference, then the Archimedes criterion is*

$$\mathrm{Ar}=\frac{gl}{v^2}\frac{\Delta T}{T}. \tag{5'}$$

From the Archimedes criterion, the following equation relating the scales of the quantities is obtained:

$$c_v=\left(\frac{c_g c_l c_{\Delta T}}{c_T}\right)^{1/2}. \tag{6}$$

Six scales enter into Eqs. (4) and (6). If one takes $c_g = 1$ (since all the experiments are carried out under earth gravity conditions), and $c_T = 1$, which makes it possible, in simulation with air, to have a model with physical constants of the medium closer to nature, only four scales remain. Hence, if one specifies two scales, the other two are determined by jointly solving Eqs. (4) and (6).

Thus, if the velocity scale is excluded from Eqs. (4) and (6), we obtain

$$c_{\Delta T}=\sqrt{\frac{c_\nu^2}{c_l}}. \tag{7}$$

Considering that admixtures of noxious substances have no substantial influence on the flow (the change of the specific weight of the mixture is considered by the Archimedes criterion), from the formula determining the discharge of the substance, we find

$$c_G=c_l^2 c_v c_q, \tag{8}$$

where c_G is the concentration scale and c_q is the scale of discharge of the noxious substances.

Thus, having created a model that is geometrically similar to nature, and maintained the similarity of the fields of physical constants at the boundaries by means of Eqs. (4), (7) and (8), one can carry out the calculation for the model, and after conducting the experiments on it, convert the results to full scale.

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*In meteorology, the parameter $B = \frac{gH}{T}\frac{\Delta T}{v^2}$, similar to the Archimedes criterion (where H is the height, assumed equal to 2m, ΔT is the temperature difference at heights of 4 and 1 m, and v is the wind velocity at the height of 2 m), is used for the characteristic, determined by the temperature distribution, of the stability of the surface atmospheric layer.

However, the fulfillment of conditions (4) and (7) causes a complication, since it leads to a model with small scales for the speeds and large ones for the temperature difference. To simplify the simulation, regions were studied in which "partial" (limited) self-similarity of the process is observed in relation to the characteristic criteria.

The experiments showed that in the surface atmospheric layer, where the impurities are propagated, within the confines of a plant site, the turbulence is substantially determined by the conditions of flow around the buildings (flow disruption at sharp edges and formation of vortex zones at the buildings). The initial flow turbulence has little effect on the concentration distribution. Thus, self-similarity takes place relative to the Kármán criterion.

In a model study of the propagation of impurities over the grounds surrounding a plant, the process is investigated in a substantially larger space (in both area and height). Perturbations caused by flow around the buildings have no substantial effect on the flow in this volume. The turbulence in the volume studied will depend considerably on the initial turbulence of the incoming flow. In this case, the process will not be self-similar with respect to the Kármán criterion. Even if a situation (topography, structure, etc.) similar to nature is created on the windward side of the model at a substantial distance (of the order of 4-6 heights of the space studied), similarity conditions cannot always be produced in this case either. In such cases, in the air flow incident on the model, it is necessary to create artificially a turbulence corresponding to the natural turbulence, and to maintain it at the necessary level over the entire length of the test section of the wind tunnel.

It has been established experimentally [5] that for low values of the Archimedes criterion, the flow may be considered isothermal with a sufficient degree of accuracy. The literature [1, 5] indicates that in the propagation of nonisothermal plumes in a stationary medium, the distortion of the plume path caused by gravitational forces may be neglected if the criterion* $Ar < 0.005$.

For nonisothermal flow of a plume in a stream, Yu. V. Ivanov [3] proposes, as the parameter characterizing the similarity, the ratio of the velocity heads in the flow and in the plume $\frac{v^2\rho}{v_0^2\rho_0}$, where v and ρ are the velocity and density of the flow and v_0 and ρ_0 are the velocity and density of the plume leaving the stack. This parameter may be used to find the boundary of the zone of "partial" self-similarity.

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*In this case, the criterion is determined by assuming the characteristic dimension (stack diameter), average exit speed of air from the stack v_0, and the temperature difference ΔT between the air leaving the stack and the air in the surrounding medium.

As was shown by experiments with two models of different scales, when the magnitude of the product is $\mathrm{Ar}\sqrt{\frac{v^2\rho}{v_0^2\rho_0}} < 0{,}05$, the presence of nonisothermality may be neglected with an error of not more than 12% (and mainly in the direction of higher concentrations in a model of smaller scale).

In the presence of self-similarity with respect to the criteria $\frac{vl}{k}$ and Ar, only the fulfillment of geometric similarity is required.

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V. M. ELTERMAN

ON EXPERIMENT OF MODELLING OF NOXIOUS SUBSTANCE DISPERSION IN THE SURFACE AIR LAYER OVER PLANT TERRITORIES*

In the USSR Central Institute of Labour Protection scientific work is conducted on diffusion process modelling in the surface air layer. Investigations for some large industrial plants were carried out which allowed to find the best decision of enterprise planning and to determine necessary stack heights.

For evaluation of data received by modelling, similarity criteria are suggested both for the case of isothermal and non-isothermal currents in which characteristic length scale, mean velocity, and velocity of turbulent energy dissipation are taken into account. Flow regions are pointed in which automodelling exists in accordance with these criteria. In such regions observance of only geometric similarity is recommended.

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*Editor's note: The abstract is presented as given in the English with the original Russian article.

SPECIAL CASES OF VERTICAL CURRENTS

I. G. Diaconescu, M. Frimescu, I. Moroianu, and A. Moroianu
RSR (Romanian Socialist Republic)

From Glavnoe Upravlenie Gidrometeorologicheskoy Sluzhby Pri Sovete Ministrov SSSR. (Chief Administration of the Hydrometeorological Service Under the Council of Ministers of the USSR.) "Meteorologisheskie Aspekty Zagryazneniya Atmosfery". (Meteorological Aspects of Air Pollution.) Sbornik dokladov na mezhdunarodnom simpoziume v Leningrade - Iyul' 1968 g. (Reports delivered at the International Symposium in Leningrad - July 1968.) Pod redaktsiey d-ra fiz.-mat. nauk M. E. Berlyanda. (Edited by Prof. M. E. Berlyand.) Gidrometeorolgicheskoe izdatel'stvo, Leningrad, p. 362-366, (1971). (Hydrometeorological Publishing House, Leningrad, (1971).)

The investigation of air pollution with emissions of industrial plants consists primarily in the study of the patterns of distribution of pollutants with time and in the space around the sources. A number of computational formulas have been derived for this purpose [1, 8, 11].

It is well known that the propagation in the atmosphere of pollutants discharged by industrial plants depends on different, simultaneously operating factors: characteristics of the emission (amount of pollutants discharged, their nature and physicochemical properties, and stack height of parameters that can be determined in advance), orography and roughness of the terrain (parameters determined experimentally), wind and atmospheric turbulence (parameters determined experimentally and theoretically).

Consideration of all these factors operating simultaneously introduces certain difficulties into the theoretical treatment of turbulent diffusion of noxious substances, and for this reason some of the solutions obtained are based on simplified hypotheses. Primarily, they exclude the distorting effects caused by irregularities of the relief and study simplified models of air motion [5, 3].

In conformity with the above, various authors, including O. Sutton [9-11], F. Pasquill [8], M. Ye. Berlyand [1, 2] and others obtained some definite results.

In the general case, turbulent diffusion is described by the equation

$$u\frac{dq}{dx} - w\frac{dq}{dz} = \frac{d}{dz}\left(k_z\frac{dq}{dz}\right) + k_y\frac{d^2q}{dy^2}. \qquad (1)$$

where q is the impurity concentration, u is the wind velocity, w is the vertical rate of deposition of the impurity, k_z and k_y are the corresponding coefficients of turbulence, the x axis is parallel to the mean wind, the y axis is perpendicular to the x axis in the horizontal plane, and the z axis is directed along the vertical.

Thus, in addition to the wind velocity (u), frequently assumed to be constant, atmospheric diffusion is acted upon by the vertical rate of deposition of the impurity and also by the coefficients of turbulence k_z and k_y, i.e., parameters whose values and variations are usually the subject of hypotheses.

For a dissected relief of the terrain, the vertical motions change substantially and play a significant role in turbulent diffusion.

The determination of the vertical motions of air is usually carried out by means of classical methods (pilot-balloon sounding by means of two theodolites). We processed the data obtained by using methods that permitted us to carry out the processing in a comparatively short time.

The first method consists in processing the sounding data by means of a specially constructed slide rule. The values indicated on the slide rule are logarithms of trigonometric functions and of the numbers 1 to 10. By using this slide rule, one can obtain the distance from the point of takeoff of the balloon to its projection on the horizontal plane and the coordinates of the projection point and of the balloon heights. The time variation of these elements is determined graphically.

The second graphical method, although less accurate, was used to obtain the data rapidly, this being convenient under expeditionary conditions. This method consists in repeating, on a horizontal plane, the motions of two theodolites tracking the balloon by using two angle gauges located in the desired scale at a distance equal to the length of the base of the theodolites. The angle gauges have a slit along the length of the ray, the slit passing through the division of 0°. The position of the balloon, projected on the horizontal plane, is noted at the intersection of two slits. The projection is made on tracing paper attached to graph paper, on which one reads off the distance from the launching site to the balloon projection point, the balloon height at different times, and then the wind direction and velocity.

The air current in valleys or across ridges is caused by horizontal gradients of pressure and temperature in the atmosphere and also by the configuration of the relief, which includes additional components [4].

A special role in the propagation of impurities is played by mountain-and-valley wind. Changes in the direction of a mountain-and-valley wind which take place in the morning and evening exert an influence on the mixing of impurities and their motion which is not considered in Eq. (1). Observations made in a valley make it possible to distinguish the direction of the air currents and also the fluctuations in the wind rotation level.

The characteristics of mountain-and-valley wind are substantially determined by the degree of dynamic turbulence caused by the relief and dependent on the wind velocity and wind direction relative to the ridge and valley.

Thus, vertical motions assume a complex character (Fig. 1). By making observations in the same valley with the aid of two theodolites in order to determine the vertical motions, we detected eddy currents with a horizontal axis. Thus, on 12 October 1966 at 8 A.M. in fair weather, the balloon was lifted by ascending current to a height of 1900 m, and then sharply dropped to a height of 900 m in 1 min. The eddy lasted until noon, slowly diminishing and gradually descending toward the ground, so that in 4 hours it reached the zone between 200 and 100 m. In their stability and intensity in the mountain zone, such eddies are of special interest to aviation. Let us note, however, that such turbulent motions play a decisive role in the propagation of impurities.

The indicated facts complicate a theoretical solution of the problem of atmospheric diffusion in valleys, where the wind intensity is attenuated, and the recurrence frequency of calms exceeds 50%. For this reason, the problem should be solved experimentally for each specific case.

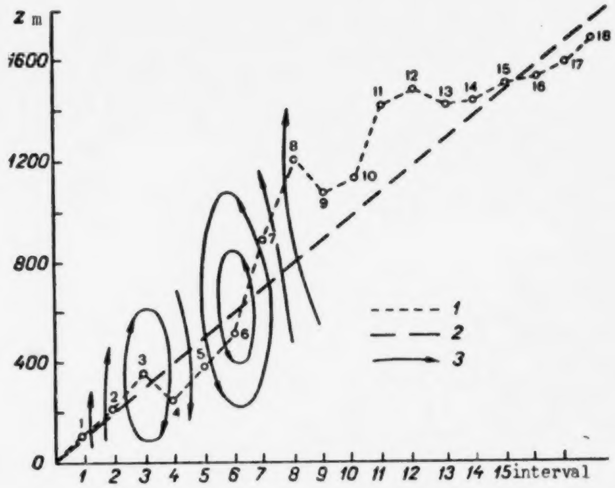

Fig. 1. Vertical motions of air in the mountain zone (8 A.M., 11 October 1966).
1 - experimental curve, 2 - theoretical curve, 3 - air currents.

On flat terrain, the effect of dynamic turbulence virtually disappears. For this reason, here one can successfully use equations of type (1). However, in these zones it is important to consider processes of thermal origin caused by the influence of differences in the underlying surface (vegetative cover and particularly, surface of the water).

In order to study the vertical currents near the Danube, observations were made simultaneously near the river and at a distance of about 10 km (with a level difference of approximately 90 m). It was thus found that descending currents exist above the Danube. At the same time, above dry land, ascending currents arise which together with the descending ones produce a barrier zone that may strongly affect the turbulent diffusion of noxious substances. On the basis of observations carried out in the course of 20 days during the period from April to October 1967, it was found that such a barrier zone near the water extends over 300-400 m.

Because of the daily heating of dry land in the morning during the warm period of the year, ascending currents are formed that encompass the surface air layer and extend to a height of 300 m, where they encounter the descending currents of convective origin, usually observed above a water surface (Fig. 2). The position of the barrier zone in these places is responsible for a special type of propagation of impurities. The latter rise from the surface and collect around the barrier, then are transported at this level along the direction of the wind.

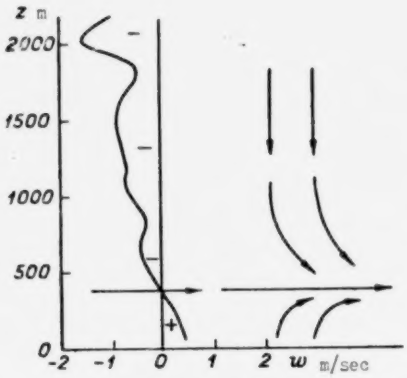

Fig. 2. Average velocity of vertical currents near the water.

The ascending and descending currents corresponding to morning convection differ around the Danube from the motions at a distance of 10 km from this river. Occasionally, descending currents in the layer from 6 to 700 m thick with speeds of over 7 m/sec were observed near the water.

Thus, by using operative methods of determination of vertical currents, we distinguished characteristics typical of mountain zones and zones near a water surface. Intense eddy currents dangerous for aerial navigation and playing an important part in the propagation of impurities around sources are observed in the mountain zone. Near the river, descending currents were observed, and also horizontal barrier zones at a height of 300-400 m, which should have a substantial influence on the propagation of noxious substances.

The study of vertical currents together with the determination of impurity concentrations should be an essential component of experimental studies of the determination of noxious substances in the vicinity of sources and particularly in zones of nonuniform relief. The nature of the underlying surface and the ruggedness of the relief act on the dynamic and thermal turbulence, and hence, on the distribution of impurities around the source, complicating the use of classical formulas of turbulent diffusion. In zones of dissected relief, it is necessary to determine the directions experimentally or to solve the problem by methods applicable to each individual case.

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I. G. DIACONESCU, M. FRIMESCU,
I. MOROIANU, A. MOROIANU

PARTICULAR CASES OF VERTICAL CURRENTS *

Observation results of vertical air motion peculiarities in mountainous region and over some parts of underlying surface with sharply different characteristics (land-water) are stated which are interesting from the point of view of their influence on pollutant distribution. Basic method of pilot-balloon observations was used for vertical motion measurement. Observations of mountainous valley found the presence of stable eddy current with horizontal axis marked between levels of 200 and 1900—1000m during four hours. Simultaneous observations near the Danube and at the distance of 10km showed that downcurrents exist over it. Up-currents covering surface air layer and distributing up to the height of 300—400m appear over the land in the morning and at noon in the warm season. At this height they meet downcurrents near water surface forming a peculiar obstruction zone preventing pollutant distribution above this level. Downcurrents in the layer with thickness of 6—700m and velocities more than 7 m/sec were sometimes observed near the water surface.

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* Editor's note: The abstract is presented as given in English with the original Russian article.

SYNOPTIC CONDITIONS OF FORMATION OF A VERY STABLE ATMOSPHERIC BOUNDARY LAYER

F. Rein CSSR (Czechoslovak Socialist Republic)

From Glavnoe Upravlenie Gidrometeorologicheskoy Sluzhby Pri Sovete Ministrov SSSR. (Chief Administration of the Hydrometeorological Service Under the Council of Ministers of the USSR.) "Meteorologisheskie Aspekty Zagryazneniya Atmosfery". (Meteorological Aspects of Air Pollution.) Sbornik dokladov na mezhdunarodnom simpoziume v Leningrade - Iyul' 1968 g. (Reports delivered at the International Symposium in Leningrad - July 1968.) Pod redaktsiey d-ra fiz.-mat. nauk M. E. Berlyanda. (Edited by Prof. M. E. Berlyand.) Gidrometeorolgicheskoe izdatel'stvo, Leningrad, p. 367-371, (1971). (Hydrometeorological Publishing House, Leningrad, (1971).)

As we know, air pollution in industrial regions depends considerably on the thermal stability of the atmospheric boundary layer. In this connection, it is of interest to consider cases of development of a very stable atmospheric boundary layer of large vertical thickness.

The present study deals with the synoptic conditions of emergence of such cases in the nonuniform landscape of the northwestern part of Czechoslovakia.

A very stable boundary layer may be characterized with the aid of the mean temperature lapse rate, whose magnitude does not exceed +0.4°C./100 m, i.e., all the inversions and isothermies together with small positive values of the temperature lapse rate are included. As far as the characteristics of the structure in height are concerned, two variants have been proposed on the basis of empirical treatments: (a) the lower boundary of a very stable layer coincides with the earth's surface, and (b) the lower boundary of such a layer is at a certain height (of the order of 10-100 m) above the earth's surface, above a layer with a high positive lapse rate.

The wind velocity at the upper boundary of a stable boundary layer is characterized by frequent and abrupt increases.

The above results in an empirical model of the possible structure of a very stable boundary layer, shown in Fig. 1. One can distinguish two types of vertical temperature distribution under the upper boundary of the boundary layer (a very stable stratification from the earth's surface or only from a certain height above the surface) together with two independent velocity profiles (a slight or sharp increase in velocity near the upper boundary of the boundary layer).

Empirically established types of a stable boundary layer were studied on climatological data for the northwestern region of Czechoslovakia. This region is located in an area with a nonuniform hilly relief and contains a concentration of many industrial plants as a result of which the air pollution reaches high proportions.

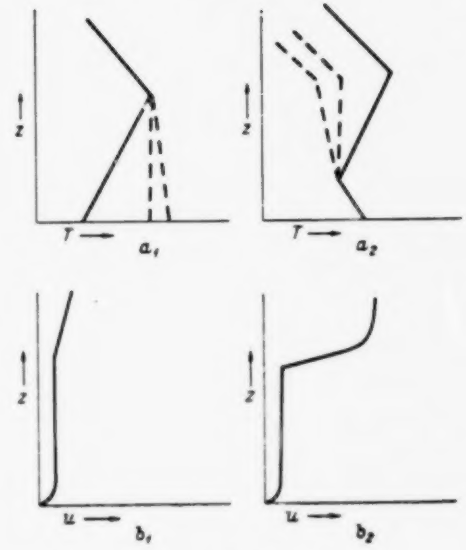

Fig. 1

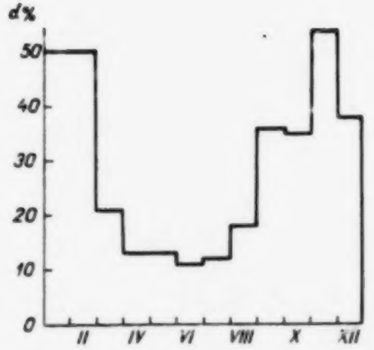

Fig. 2

To determine the frequency of cases of a very stable boundary layer with a thickness of the order of several hundred meters above the surface, we used data from aerological observations in this region, supplementing them with temperature measurements from meteorological stations in the mountains and valleys.

Based on data of climatological treatment of a five-year series of measurements (1956-60), the following results were obtained: a group of cases with a very stable boundary layer was observed for an average of 29% of the days per year, and the frequency is marked by a sharply defined annual variation (Fig. 2). Of this number, a 75% frequency is reached by cases denoted by a_1 in Fig. 1, i.e., those involving a very stable stratification directly from the earth's surface. In the distribution of cases from the standpoint of the wind profile (based on measurements of stations in the valleys and mountains), the frequency is 53% for group b_1 and 47% for group b_2 (groups a_1 and a_2 being independent of groups b_1 and b_2).

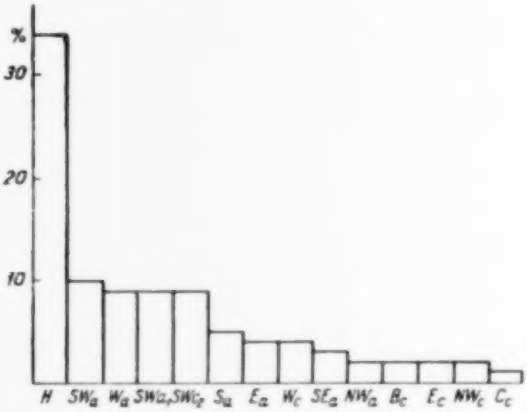

Fig. 3

Thus, from climatic data one can determine that the empirical groups a_1, a_2, b_1, and b_2 of a very stable boundary layer are characteristic of this region. We will later try to find their relationships to synoptic situations.

First of all, the question arises, in what synoptic situations can one expect the formation and development of a vertically thick, very stable boundary layer in the landscape studied. The general answer is supplied by Fig. 3, which shows the relative frequencies of days with stable boundary layers of the total number of days with individual synoptic situations according to the classification of Koncek and the author.

It is evident from Fig. 3 that a very stable boundary layer arises relatively often in the presence of the Middle European anticyclone (H), i.e., in 34% of the days when an anticyclone is observed.

The second frequency group is formed by synoptic types with a southern component of advection, i.e., the southwestern anticyclonic (SW_a) and cyclonic (SW_{c_1} SW_{c_2}) types, together with the southern and southeastern anticyclonic types (S_a - SE_a). The remaining cases are of no particular interest from the standpoint of their frequency.

If this result is compared with the frequency of appearance of dense fogs in the lowland of the region studied, a good agreement of the advection values will be noted. Most fogs are observed in calms and in advection with a southern component, practically no fogs being observed in synoptic situations with a northern component. This phenomenon is apparently due to the fact that northern winds descending from the mountain ridge with a considerable turbulence do not promote the development of a thick, very stable boundary layer.

The next objective was to study the dependence of individual types of a very stable boundary layer (a_1, a_2, b_1, b_2) on the synoptic situations.

The frequency distribution of groups a and a_2 is as follows: the first constitutes 75% of all cases of a very stable boundary layer and appears primarily in anticyclonic situations. Group a_2 is observed in 25% of the cases and mostly in cyclonic situations.

Groups b_1 and b_2 are distributed in the manner of groups a_1 and a_2. Group b_1 is observed in 73% of the cases, appearing primarily in the central anticyclone and in the remaining synoptic situations with a slight tropospheric advection. Cases b_2, which account for 23%, arise when advection predominates, i.e., on the outskirts of baric formations.

Thus, this paper has attempted to establish a relationship between the very stable boundary layer in the northwestern part of Czechoslovakia and synoptic situations.

The boundary layer, which reaches an altitude of several hundred meters above the surface and is practically uniform over a distance of tens of kilometers, is a mesometeorological object related to the action of local radiation factors and to the influence of macrocirculation fields.

From this point of view it is clear that a stable boundary layer depends on anticyclonic situations. However, in about one-quarter of the cases, a very stable boundary layer is observed in this area in cyclonic situations. Such a situation arises mainly when anticyclones alternate with cyclones with such a direction of advection that this area is not situated on the lee side of Krusne Mountains.

Work on details of the structure of the boundary layer in the investigated region is continuing.

F. REIN

SYNOPTIC CONDITIONS OF APPEARANCE OF VERY STABLE BOUNDARY AIR LAYER*

The investigation results of occurrence frequency of powerful and very stable boundary layer (mean lapse rate $< 0.4°/100m$) are stated in dependence on synoptic conditions for nothern-western part of Czechoslovakia, distinguished by hilly relief and high level of air pollution by industrial plant emissions. With the help of climatological treatment of aerological observation series for 5-years it has been found that such a layer can be met in 29% of days a year, its frequency having distinctly expressed annual course with minimum in summer and maximum in winter. Very stable boundary layer arises most often in the presence of Middle-European anticyclone (34% of days in which anticyclone is observed). Synoptic situations with southern advection component form the second of frequency group, out of which 25% of cases refer to cyclonic situation. It is marked that in 75% of cases observed chiefly in anticyclonic conditions the lower boundary of a layer with small lapse rate coincides with underlying surface, and sharp increase of wind velocity occurs at the upper boundary.

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* Editor's note: The abstract is presented as given in English with the original Russian article.